Do these residues sent from Rockwell-Hanford at Richland, Washington contain salvagable plutonium? Members of LASL's Plutonium Facility determine the kinds and amounts of special nuclear materials in such heterogeneous waste with nondestructive assay techniques (the segmented gamma scanner and the thermal neutron coincidence counter). Measurements of this sample indicated that the contents were not worth recovering.

Nondestructive Assay for Nuclear Safeguards

by Roddy B. Walton and Howard O. Menlove

Sophisticated nondestructive assay techniques have solved many difficult measurement problems in the nuclear fuel cycle, but their potential for keeping track of sensitive nuclear materials on a nearly continual basis is just beginning to be exploited in nuclear facilities.

Radioactive nuclear materials reveal their presence by the radiation they produce spontaneously or otherwise. These characteristic signatures form the basis for nondestructive assay (NDA) of sensitive nuclear materials and for modern safeguards measurement technology. NDA techniques are used now by the nuclear industry, defense facilities, and safeguards inspectors to make rapid, accurate measurements of sensitive nuclear materials in diverse forms and compositions and thus to close many gaps in inventory measurements. However, this technology has not drastically changed the overall materials accounting practices of most nuclear facilities. Instead, the nuclear industry has placed heavy emphasis on physical protection to safeguard nuclear materials against overt threats of diversion. Further, a major part of the National Safeguards Program is devoted to increasing these physical protection measures.

The Los Alamos Scientific Laboratory (LASL) program, on the other hand, has pioneered the development of NDA technology needed not only for inventory closure but also for near-real-time measurement and accountability systems. NDA techniques can be used to assure that no sensitive nuclear materials (SNM) are being lost or diverted from their defined flows and containments. Rapid measurements of SNM in feed materials, process lines, finished products, scrap and waste, and holdup in the plant are all possible with NDA techniques. When put together in an integrated materials control and accountability system, they can deter protracted diversion of SNM by a knowledgeable insider by detecting the amount and location of losses in a timely fashion. Such systems can also determine the validity of threats that significant amounts of nuclear material have been diverted for unauthorized purposes.

At present several NDA-based near-real-time materials accounting systems are in existence and we are helping to plan such systems for future high throughput fuel production and spent-fuel reprocessing facilities. We also continue to work closely with members of the nuclear industry and safeguards inspectors to solve individual measurement problems associated with accurate inventory measurements, quality assurance of nuclear fuel production, and adequate safeguards inspection procedures. For many existing facilities, accounting systems that combine conventional chemical analysis with NDA techniques can provide adequate safeguards, but advanced facilities will require upgraded approaches.

Uranium and plutonium are the principal raw materials of fission weapons and hence the most sensitive nuclear materials to safeguard and to measure.


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Moreover, because their fissile content or isotopic composition determines their strategic value as well as their value for nuclear reactor applications, it is important to measure isotopic compositions as well as absolute amounts of these elements. For example, natural uranium contains 0.7% of the fissile isotope ^{235}U, with the remainder being the fertile isotope ^{238}U. Both fissile and fertile isotopes will fission when bombarded with neutrons, but fissile isotopes are the important ingredient in nuclear fuels because they fission with high probability when bombarded by low-energy neutrons, whereas fertile isotopes will not fission unless they are bombarded by high-energy neutrons. Plutonium-239 is the principal fissile isotope in all plutonium fuels. This isotope is produced in nuclear reactors through the reaction

$$^{238}U + n \rightarrow {}^{239}U \rightarrow {}^{239}Np + \beta + \gamma\text{'s} \rightarrow {}^{239}Pu + \beta + \gamma\text{'s}$$

Uranium-238 is called fertile because it produces the fissile isotope ^{239}Pu by neutron capture. Other fertile isotopes are ^{240}Pu, which produces fissile ^{241}Pu by neutron capture, and ^{232}Th, which produces ^{233}U. Thus safeguards measurements are concerned with determining the presence of a large number of fissile and fertile isotopes of uranium, plutonium, and thorium.

In this article we discuss nuclear NDA methods based on detecting either the natural radioactivity of these materials (passive assay) or the radiations produced when these materials are bombarded by external sources of neutrons and gamma rays (active assay). The methods are rapid, usually requiring only a few minutes to complete a measurement. They are nondestructive in the sense that the materials can be measured without removing them from their containers and that the measurement is based on observation of the decay or transmutation of a negligible number of nuclei relative to one mole. Moreover, they interrogate the entire bulk of the material rather than just a sample and are therefore capable of accurately measuring heterogeneous materials such as scrap and wastes from processing.

One of the earliest, most critical needs for NDA methods was for the measurement of nuclear process scrap and waste. Chemical analysis was not reliable because of the problem and expense of obtaining representative samples from these characteristically heterogeneous materials. Some large inventory discrepancies (for example, the problem encountered at the NUMEC facility in Apollo, Pennsylvania, in 1965) have been difficult to resolve because of deficiencies in such measurements. Good measurements of scrap also were needed to assess the material's value for recycle, and NDA methods for the screening and assay of low-level waste were needed to dispose of the materials safely and economically.

NDA techniques were also required to extend the ability of safeguards inspectors to check facility operator compliance with safeguards requirements—in particular, to check that measured material inventories matched declared "book" inventories within the limits of measurement uncertainties. To accomplish this objective, inspectors carry out detailed auditing of records and procedures; perform on-the-spot inventories of containers of nuclear materials, which involve weighing and verifying seals; collect random samples for chemical analysis; and examine analytical laboratory procedures. With portable and in-plant NDA instrumentation, together with certified standards, an inspector can independently verify the types and amounts of materials in a facility, including materials held up in process lines.

International Atomic Energy Agency (IAEA) inspectors present special challenges for measurement instrumentation because they work under difficult political and physical constraints. The terms of the safeguards agreements between the country being inspected and the IAEA define the political constraints. For example, their terms limit materials accessibility and inspection time. Further, the inspector must work in a foreign environment without the normal supporting services, such as calibration standards, that are available for domestic safeguards. Equipment therefore must be lightweight, reliable, rugged, and easy to calibrate.

The LASL Safeguards Program has studied and continues to study these and other challenging measurement problems. In most cases, the problems have been identified by domestic and IAEA safeguards inspectors, Department of Energy (DOE) field offices, DOE facility operators, DOE safeguards systems analysts, and designers of integrated safeguards systems for new fuel cycle facilities, as well as the operators of LASL facilities, principally CMB Division staff. Because we want to gain widespread acceptance of our new technology, we consider test and evaluation of a fully engineered prototype instrument in the operating environment of a host nuclear facility to be the most important phase of the development of a measurement method. We also prepare comprehensive design and performance documentation to facilitate the work of instrumentation vendors and potential users.

In the remainder of this article we describe (a) techniques for nondestructive assay of fissionable material including some applications of major methods, (b) the status of NDA technology development and its implementation in the fuel cycle and DOE facilities, and (c) some future challenges in this field of measurement technology. Many laboratories over the world have

Fig. 1. Examples of containers used for nuclear materials in fuel cycle facilities. The large containers are used for low-concentration scrap and waste, the intermediate-size containers (1- and 4-L) hold high-purity process fuel materials or high-concentration scrap, and the small vial is a typical container for samples withdrawn from process lines for chemical analysis.

contributed significantly to the development of this technology, but rather than present a comprehensive review, we will focus on LASL-developed instrumentation and methods.

Characteristics of a Nondestructive Assay System

No single NDA instrument or method will suffice for the assay of the diverse forms and containments of nuclear materials in the fuel cycle. Some typical samples are shown in Fig. 1. Typical "feed" materials for input to the fuel fabrication process are UF_6 in metal cylinders with capacities for up to $\sim 10^4$ kg, depending on enrichment; plutonium nitrate solution in 10-L plastic bottles (120 cm long); and uniform fuel blends in few-liter cans. Reactor fuel materials include pellets, plates, rods, and bundles. Recoverable scrap, such as defective product material and calcined process residues, is stored in few-liter containers. Slightly contaminated wastes, such as paper, rags, and rubber gloves, are often stored in 120- and 220-L drums.

Clearly then, the capability to "view" bulk materials quantitatively is essential for most applications; hence, for both active and passive assay the interrogating beams and radiation signatures of the nuclear materials being measured must be highly penetrating. Neutrons and gamma rays are obvious choices from the domain of low-energy nuclear physics, as opposed to less penetrating alpha and beta rays. Additionally, calorimetry, which is based on the measurement of heat generated by radioactivity, can be used to measure plutonium with high precision, provided the isotopic composition of the plutonium is known from independent mass spectrometry or gamma-ray spectrometry measurements. (Uranium does not generate enough heat to be measured with calorimetry.)

The characteristics that are common to all practical NDA systems are:

1. an assay principle based on the detection of suitably copious fundamental signatures of the isotopes or elements to be measured, that is, neutrons or gamma rays from natural radioactivity or from interrogation with an external source;
2. a detection system optimized for the signature selected and the types of samples to be measured;
3. electronic and mechanical systems for data acquisition and assay controls; and
4. a consistent means for obtaining the desired mass of the isotope or element from observed counting data.

Detectors for NDA instruments are essentially solids or gases that ionize as they interact with incoming neutrons or gamma rays. The amount of ionization is proportional to the energy deposited in the detector by each interaction. The generated charge forms an electronic pulse that is then amplified and processed and either counted or analyzed by electronic and microprocessor units. Gamma-ray detectors used in NDA instruments are solid crystals of NaI and germanium, standard tools of the gamma-ray spectroscopist. In NaI detectors, the ionization in the crystal produces light (scintillation), which is converted to an electronic pulse by a photomultiplier tube. In germanium and other solid-state detectors, the charge produced is converted directly into an electronic pulse. The neutron detectors most commonly used for NDA instruments are gas proportional counters, filled with ^3He, BF_3, or ^4He; however, plastic or liquid scintillators also can be used.

The conversion of counting-rate data to mass determinations or isotopic abundances of SNM involves the most subtle and difficult problems. Among these are calibration of the instruments and corrections for absorption and scattering of the signature radiation by the sample being measured. The sample includes not only the particular isotope or element of interest, but also "matrix" materials in which the material of in-

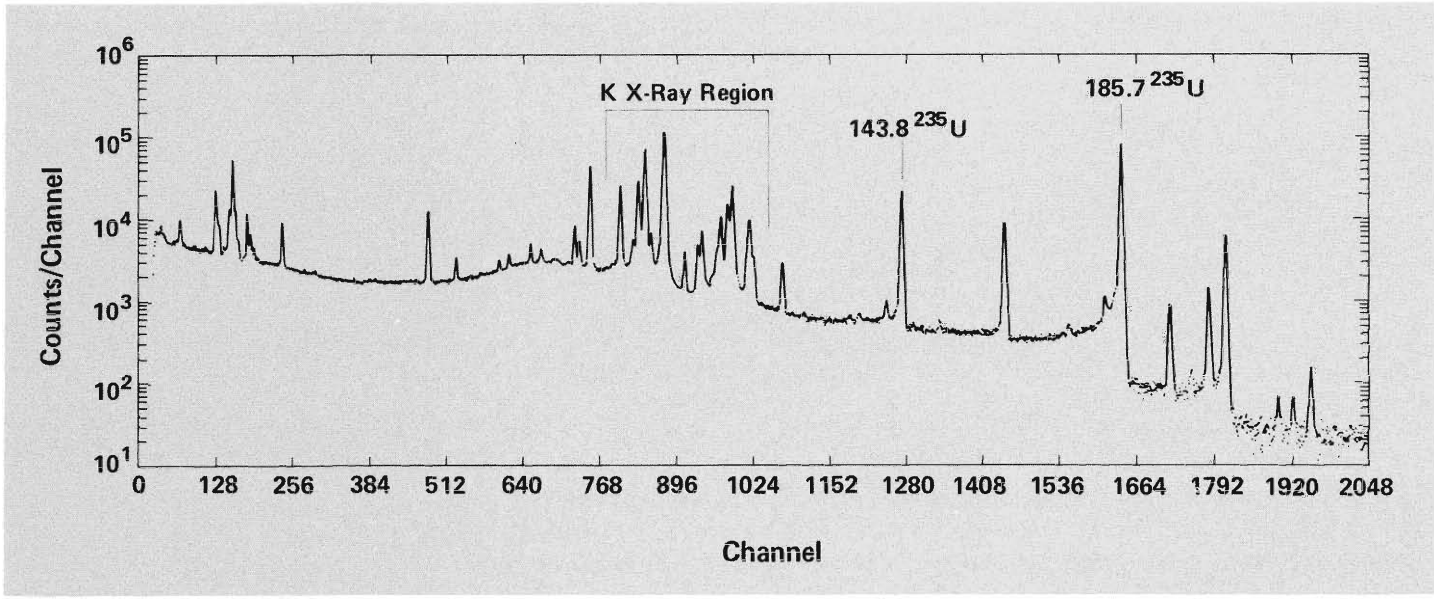


Fig. 2. Pulse-height distribution of gamma rays from high-enriched (93% ^{235}U) uranium, measured with a high-resolution germanium detector. Gamma-ray energies (in keV) are noted on some of the peaks. The prominent peak at 186 keV is used for passive assay of ^{235}U.

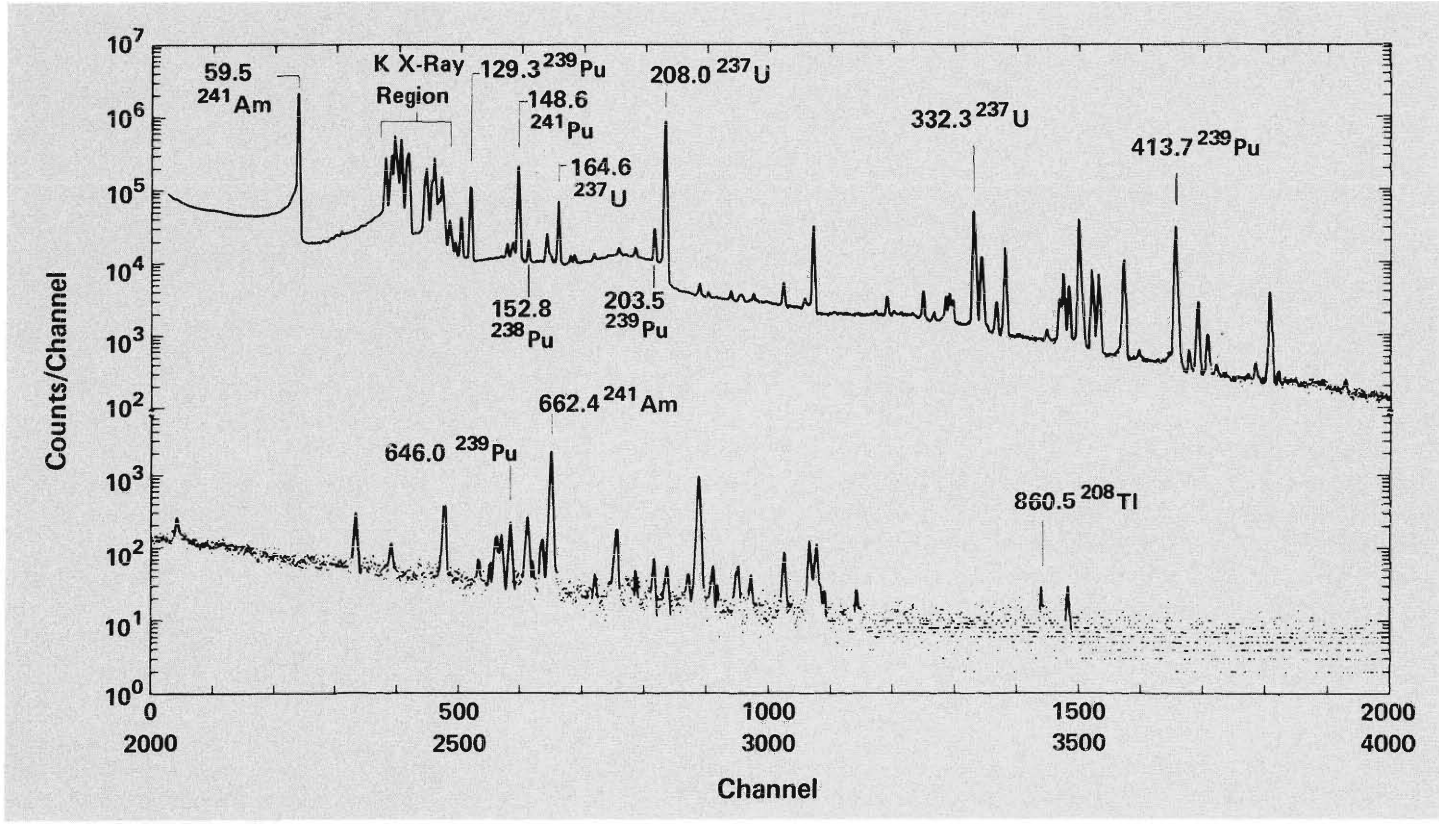


Fig. 3. Pulse-height distribution of reactor-grade plutonium (15% ^{240}Pu), measured with a high-resolution gamma-ray detector. Gamma-ray energies (in keV) are noted on some of the peaks.

terest is embedded. In an active method, one also has to account for the absorption and scattering of the interrogating beam of photons or neutrons as it penetrates the sample. If the chemical and isotopic compositions of samples are well known, as with identical cans of PuO_2 powder, then the correction problem may be circumvented simply by calibrating the measurements with a set of standards that closely cover the range of unknowns. However, we prefer a method whose response is as independent of sample matrix materials as possible, to reduce the number of calibration standards. It is even more important to reduce matrix dependence in measurements of poorly characterized scrap and waste materials. We use the sophisticated radiation transport codes developed over the years for defense projects and reactor design to guide the design of an assay system.

General Methods for Passive Gamma-Ray Assay

Each isotope of SNM is radioactive and decays at a known rate through alpha or beta decay (for example, $^{235}U \rightarrow {}^{231}Th + \alpha + \gamma$'s and $^{239}Pu \rightarrow {}^{235}U + \alpha + \gamma$'s). These spontaneous decays produce a characteristic spectrum of gamma rays, specific in both quantity and energy. Thus a direct measurement of the amount of SNM in a sample can be obtained by counting the gamma rays it emits at specific energies.

The pulse-height spectra of gamma rays from high-enriched (93% ^{235}U) uranium and from reactor-grade (15% ^{240}Pu) plutonium are shown in Figs. 2 and 3, respectively. The isotopic abundances of the reactor-grade plutonium are: $^{238}Pu = 0.15\%$, $^{239}Pu = 81.6\%$, $^{240}Pu = 15.2\%$, $^{241}Pu = 2.4\%$, $^{242}Pu = 0.66\%$, and $^{241}Am = 0.8\%$. These data were taken with a high-resolution germanium gamma-ray detector and a multichannel pulse-height analyzer. The plutonium spectrum is very complex, with contributions from several hundred gamma rays from ^{238}Pu; ^{239}Pu; ^{240}Pu; ^{241}Pu, and the daughters or products of ^{241}Pu decay; ^{237}U; and ^{241}Am. (In Fig. 3, a few of the peaks are labelled with their energies and isotopic origins.) Furthermore, the gamma-ray spectrum of plutonium changes with time because of the relatively short half-lives of ^{241}Pu and ^{237}U.

An assay is based on determining the areas under one or more peaks in the spectrum, usually calculated with a minicomputer that is an integral part of a multichannel-analyzer, data-acquisition system. We use either NaI or germanium gamma-ray detectors, but we prefer germanium because its higher (30X) resolution permits cleaner separation and more accurate analysis of the gamma-ray spectrum peaks. On the other hand, for field measurements, such as the assessment of material holdup in process equipment, portable assay units comprising a NaI detector and single-channel electronic analyzers set to bracket specific peak and background regions of the gamma-ray spectrum are satisfactory.

For measurement of uranium, we use the prominent 186-keV gamma ray from ^{235}U; 4×10^4 gamma rays are emitted per gram ^{235}U per second. Similarly we use the 414-keV gamma ray from ^{239}Pu, which has a comparable intensity, to measure plutonium. To convert counting rates of these isotopes to total uranium or plutonium, their isotopic abundances must be known or measured independently. In all applications except spent-fuel recovery, uranium isotopes other than ^{235}U and ^{238}U are present in such small amounts that they can be neglected. Uranium-238 can be measured using as the signature the 1.001-MeV gamma ray from the decay of its daughter ^{234m}Pa. However, the measurement must be made at least 3 months after chemical purification of

uranium so that the daughters are in equilibrium. The isotopic abundances of plutonium are much more complex, but still they can be measured by performing a detailed analysis of gamma-ray pulse-height spectra (described below).

The simplest gamma-ray arrangement, a portable system that has found extensive application in plant surveys, is shown in Figs. 4 and 5. The assay system consists of a NaI detector housed in a lead collimator, pulse-processing electronics (contained in the small box in Fig. 5), a sample turntable, and an external gamma-ray source for determining the gamma-ray attenuation of the sample. An assay is performed by collecting counting-rate data with the unknown sample, measuring the transmission of an external beam of gamma rays through the sample, and applying a correction for the sample self-attenuation of the SNM gamma rays derived from the transmission measurement. We calibrate the system by performing measurements of known standards in the same manner.

The central problem in the NDA of bulk samples is the correction for sample self-attenuation. That is, the emitted gamma rays are scattered and absorbed within the sample itself. Attenuation is large and difficult to anticipate because the gamma rays have low energies, typically 100 to 400 keV; the samples frequently contain high-Z elements that absorb strongly; and the chemical composition of the sample is often unknown. The attenuation of gamma rays in a sample is given by $e^{-\mu\rho X}$ where ρ (g/cm^3) is the density of the medium, μ (cm^2/g) is the mass attenuation coefficient, and X(cm) is the thickness of the sample. The gamma-ray attenuation also can be written as $e^{-\mu_\ell X}$, where μ_ℓ is the linear absorption coefficient. Table I gives the mean free path, $1/\mu_\ell$, of the ^{235}U 185.7-keV and the ^{239}Pu 413.7-keV gamma rays for several materials. Note that high-Z materials cause a dramatic

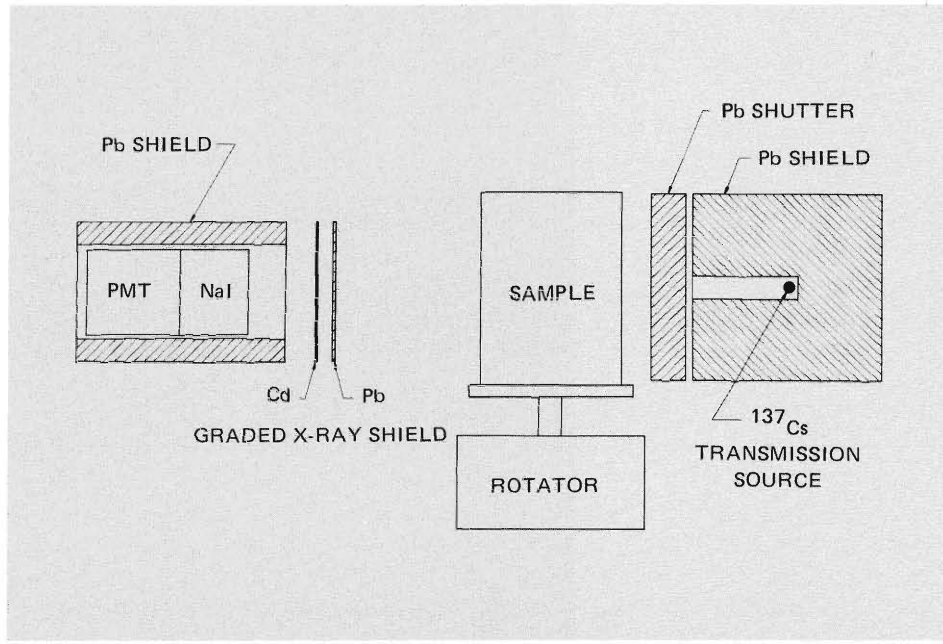


Fig. 4. A passive gamma-ray assay measurement setup. The sample is placed on a rotating table, and the detector (at the left) counts the gamma radiation from the sample. Correction for the attenuation of the SNM gamma rays by the sample is determined by measurement of transmitted gamma rays through the sample from an external radioactive source (in this case, ^{137}Cs) shown at the right.

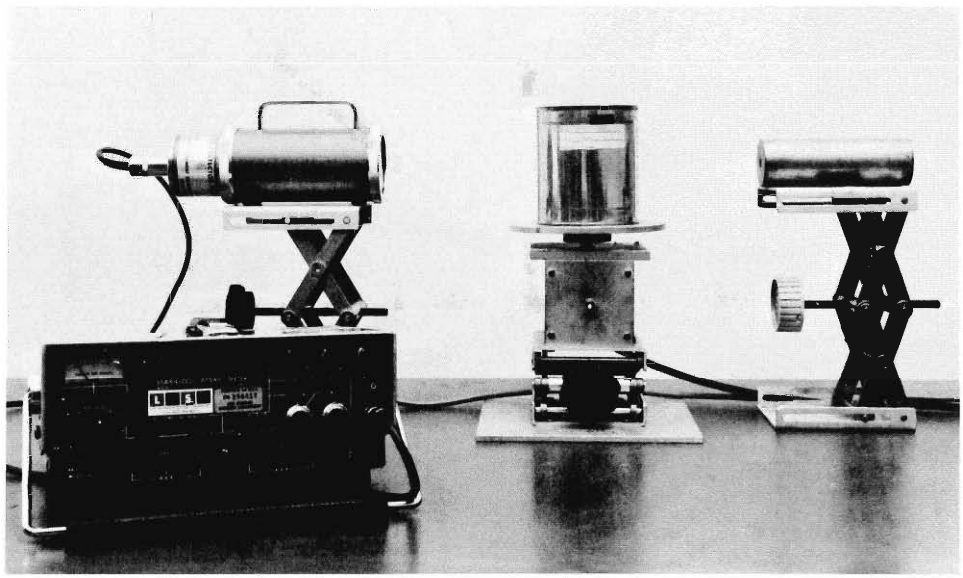


Fig. 5. A simple passive gamma-ray assay setup. At the left is the gamma-ray detector, a NaI crystal attached to a photomultiplier tube. The sample is mounted on a rotating table, and the transmission source is positioned to the right of the sample. (The arrangement is the same as that shown in Fig. 4.) The pulse processing electronics is an Eberline SAM-2 portable instrument located below the detector.

TABLE I

GAMMA-RAY MEAN FREE PATH

Medium	186 keV (^{235}U) (cm)	414 keV (^{239}Pu) (cm)
H_2O	7.1	9.7
Al	3.0	4.1
Fe	0.84	1.4
Pb	0.071	0.44
U	0.033	0.21

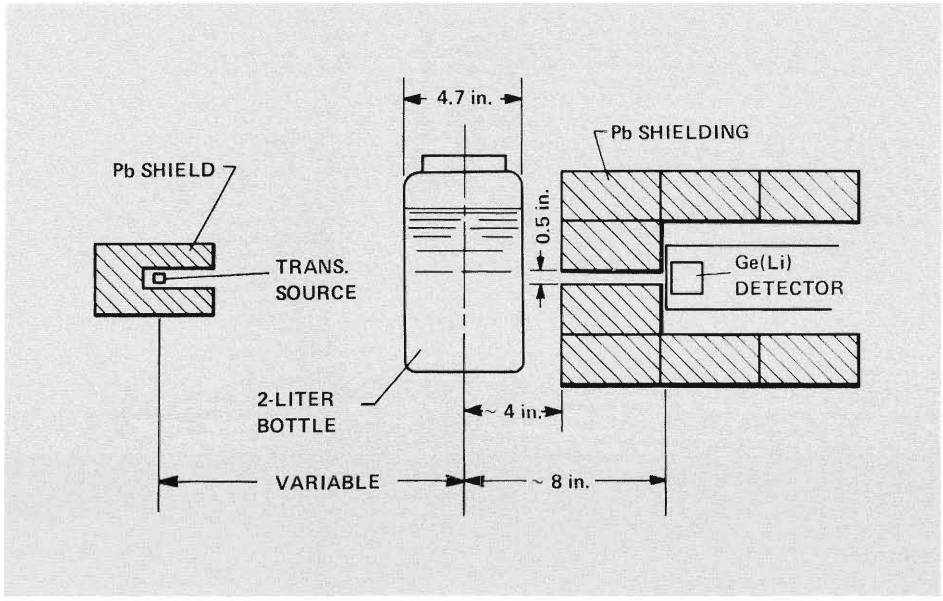


Fig. 6. The segmented gamma-ray scanning system used for passive gamma-ray measurement of heterogeneous uranium and plutonium scrap and waste. The sample is rotated and assayed by segments as it is translated vertically past the collimated germanium detector shown on the right. Transmissions of gamma rays from the source on the left are used to derive self-attenuation corrrections for each assay segment.

decrease in the gamma-ray mean free path, especially for the lower energy gamma ray. Even small particles (~0.02 mm) of metallic uranium or plutonium are highly self-absorbing.

To determine the self-attenuation correction, most gamma-ray assays employ a separate transmission measurement of the linear absorption coefficient μ_l of the sample. The external gamma-ray source must supply gamma rays with energies close to the energy of the signature gamma ray of interest. Once we know μ_l, the dimensions of the sample, and its distance from the detector, we can calculate the self-absorption correction provided the mixture of the material to be assayed and the matrix are reasonably uniform and the particles of assay material are small enough to ignore self-attenuation within individual emitting particles. Few closed forms exist for the correction factors, but often semiempirical analytical forms are sufficiently accurate. J. L. Parker and T. D. Reilly have developed analytical forms for self-attenuation corrections for most practical passive gamma-ray assay problems.

Segmented Gamma Scanner

The simple procedures described above are inadequate to measure containers of scrap and waste because they cannot take into account the vertical variations in SNM and matrix densities characteristic of these containers. Radial inhomogeneities are usually less pronounced, and their effects are substantially reduced by sample rotation. We developed the Segmented Gamma Scanner (SGS), which is both an instrument and a procedure, to improve the assay accuracy for a wide range of scrap and waste. It was also the first fully automated NDA instrument for Safeguards and is now used widely in fuel processing facilities.

The basic principle of the SGS is to divide the sample into thin horizontal segments and assay each segment independently using the transmission-corrected passive assay technique described above. After all the segments have been measured, the results are summed to give the total assay for the container. The method is shown in Fig. 6. A germanium detector views a segment of the container through an open slit, or collimator, in a lead shield. The transmission source is on the opposite side of the container, in line with the detector. For ^{235}U assay, the transmission source is ^{169}Yb. It emits 177.2-keV and 198.0-keV gamma rays, which closely bracket the 185.7-keV assay gamma ray of ^{235}U. The ^{75}Se 400.6-keV gamma rays serve for transmission measurements in ^{239}Pu assays based on detection of 414-keV gamma rays.

Computer control of SGS instruments allows automatic data acquisition, analysis, and management of all hardware. An operator places a container on the sample table, and the SGS does the rest. The SGS begins the assay sequence by positioning the sample table so that the top of the sample is just below the detector axis. Automatic controls rotate the sample continually and elevate it in discrete steps until all segments are assayed. Vertical profiles of individual segment assays and transmissions are available as data output, as well as the total SNM in the container

(Fig. 7). Typically a complete SGS assay requires 3 to 5 minutes, and accuracies lie in the 1-5% range for liter-size samples.

SGS instruments are now commercially available and are used in most major fuel cycle facilities to measure scrap and waste. Figure 8 shows an SGS instrument for 220-L and other large containers of low-level waste. We also have designed smaller units to assay containers less than 20 L.

The Enrichment Meter Principle

The end products of uranium fuel production are high-concentration, homogeneous forms of enriched uranium, such as metal, uranium oxide in powder and ceramic fuel forms, and rich uranium scraps. We have developed a very simple method to measure the ^{235}U enrichment in these materials, which is widely used in nuclear facilities not only to safeguard nuclear fuels but also to assure their quality. Called the *enrichment meter* technique, it uses to advantage the severe attenuation of the ^{235}U 186-keV gamma ray by high-Z materials. For homogeneous samples whose thickness is essentially infinite relative to the mean free path for this gamma ray, the intensity of the 186-keV gamma ray emitted will be independent of the sample thickness because only the gamma rays emitted near the surface will reach the detector. With reference to the sample-detector geometry in Fig. 9, performing the integration over the sample thickness indicated in the figure shows that for a metallic uranium sample, the counting rate (CR) of 186-keV gamma rays is given by the following expression.

$$CR = (\varepsilon I A^{235}\underline{U}/\mu_1) \propto (^{235}\underline{U}/\mu_1) ,$$

where

ε = detection efficiency,
I = intensity of 186-keV gamma ray (γ/s per g ^{235}U),
$^{235}\underline{U}$ = ^{235}U mass fraction (enrichment),

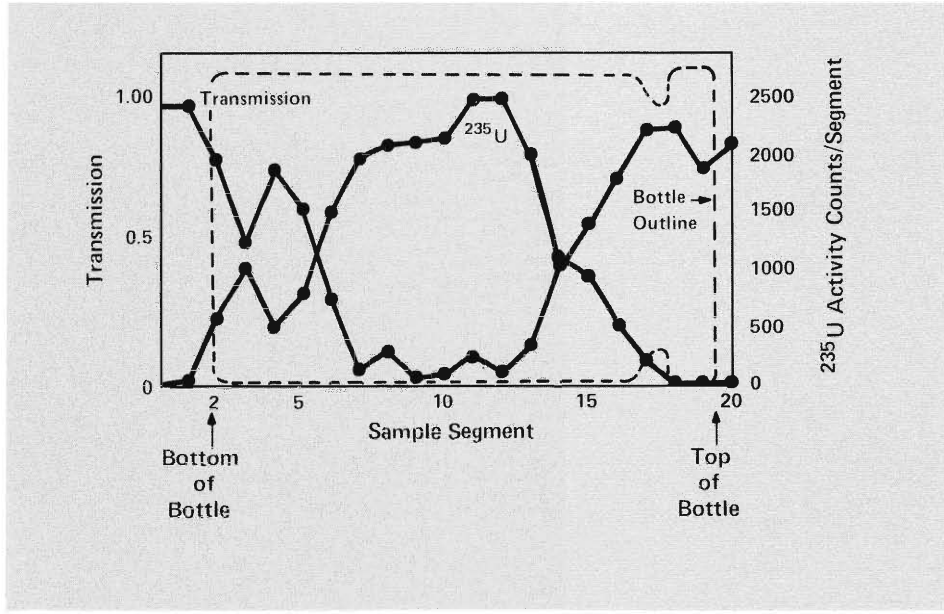


Fig. 7. Sample of data produced by segmented gamma scanner. The transmission of external rays (left-hand ordinate) and ^{235}U 186-keV gamma-ray activity (right-hand ordinate) are shown as a function of vertical segment of a 2-L bottle of high-enriched uranium. Note the profile of the bottle (lying on its side). The mirror effect in the transmission and activity data indicates that the sample attenuation is caused principally by its uranium content.

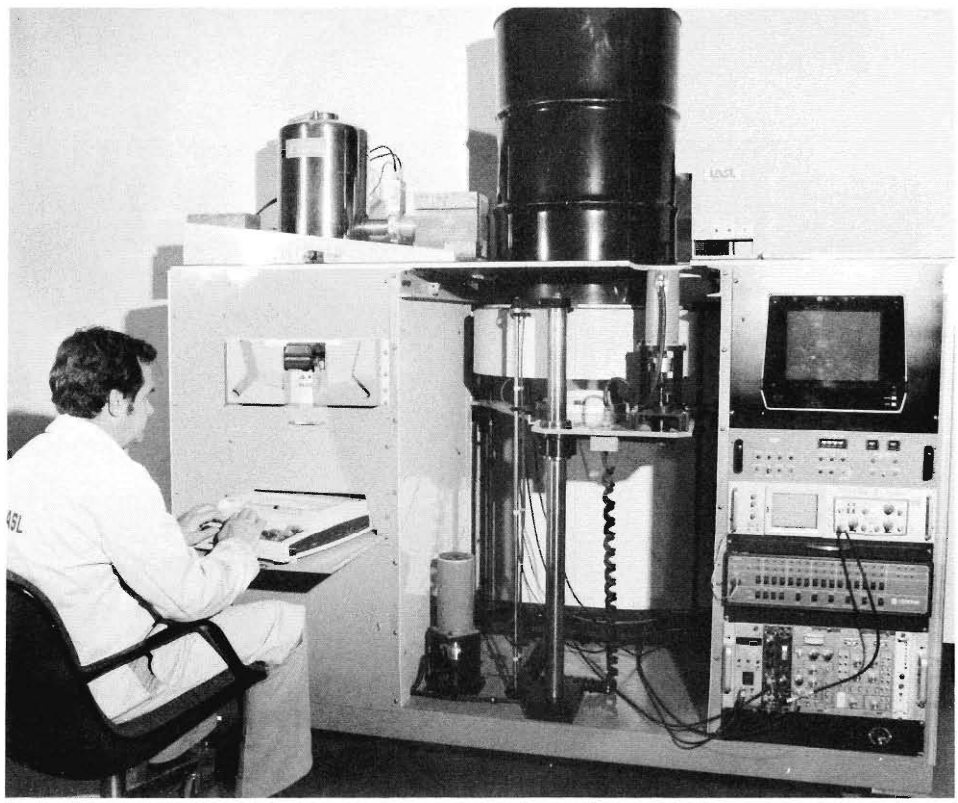


Fig. 8. An automated segmented gamma scanner for assay of uranium and plutonium in large containers. The germanium detector is located on the left of the 55-gallon drum sample, and the transmission source is on the right.

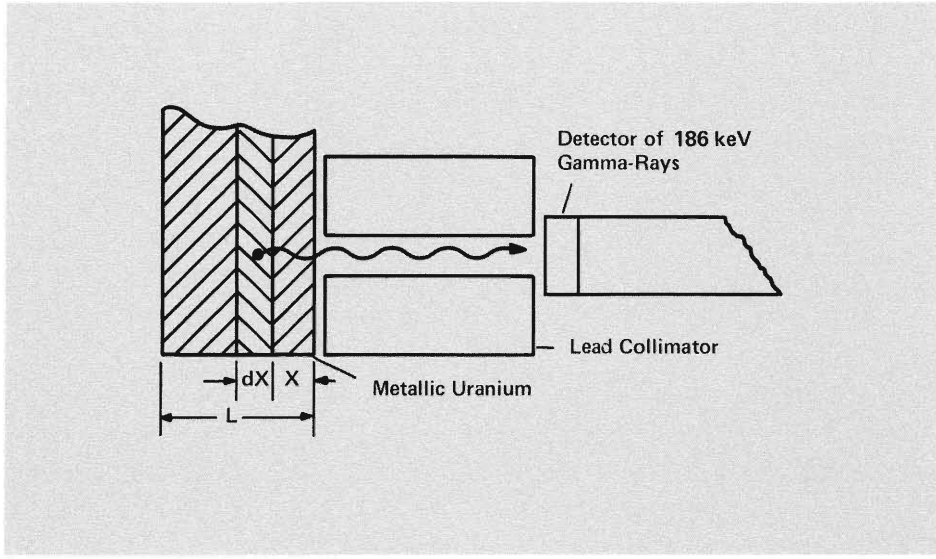


Fig. 9. The measurement geometry used for the enrichment meter principle. A measurement of 186-keV ^{235}U gamma rays from a thick high-concentration uranium sample can yield the ^{235}U enrichment directly.

A = area of sample (cm^2) defined by collimator, and

μ_1 = mass attenuation coefficient of uranium for 186-keV gammas (cm^2/g).

The counting rate is directly proportional to the ^{235}U mass fraction, or enrichment, and does not depend on sample density or other factors as long as a fixed detector and collimation geometry is maintained for calibration and measurements.

For uranium compounds such as UO_2 or UF_6, the expression for CR is

$$CR \propto (^{235}U/\mu_1)\ F\ ,$$

where

$$F = |1 + (\mu_2\rho_2/\mu_1\rho_1)|^{-1}\ ,$$

Fig. 10. Field measurement of ^{235}U enrichment of UF_6 in a 2-1/2-ton shipping container. A portable NaI detector is used to measure 186-keV ^{235}U gamma rays. A portable ultrasonic gauge is used to determine the cylinder wall thickness for the attenuation correction.

μ_2 = mass attenuation coefficient of the low-Z element, and

ρ_1, ρ_2 = the density (g/cm^3) of uranium and the low-Z element, respectively.

For matrix materials with $Z < 30$, $\mu_2/\mu_1 \leq 0.11$ and, in many practical cases, F is nearly equal to unity, being 0.988 for UO_2 and 0.930 for residues containing only 50 wt% uranium. Note also that F depends on concentration ratios instead of absolute quantities.

The enrichment meter principle may be used with a NaI or germanium detector; the latter gives more accurate results. Figure 10 shows a field measurement of the enrichment of UF_6 in a standard 2½-ton cylinder used to ship this material to light water reactor (LWR) fuel fabricators.

Plutonium Isotopic Analysis

The isotopic abundances of plutonium are needed for safeguards and accounting, particularly to distinguish weapons-grade from reactor-grade material and to assure the quality of product fuel. Furthermore, all NDA methods developed so far for the quantitative assay of plutonium in bulk materials depend on a prior knowledge of the isotopic abundances of the isotopes whose signatures are to be used for the measurement technique, that is, ^{239}Pu for gamma-ray assay based on the 414-keV gamma ray, the fissile isotopes ^{239}Pu and ^{241}Pu for active neutron methods, and all plutonium isotopes and ^{241}Am for calorimetry. In many instances, particularly for scrap and waste materials and recycle streams, the isotopic abundances of plutonium are not specified reliably.

To meet the need for a rapid nondestructive isotopic analysis, we are developing and adapting gamma-ray spectrometry. The methods involve analysis of pulse-height spectra of plutonium gamma rays measured with high-resolution germanium detectors. Peak areas are obtained by using either a simple channel summation procedure with a straight-line background subtraction or sophisticated peak-fitting algorithms. As evident from Fig. 3, the plutonium isotopes ^{238}Pu, ^{239}Pu, and ^{241}Pu, as well as the ^{241}Pu daughters ^{241}Am and ^{237}U, emit gamma rays that are useful for isotopic analysis. Unfortunately, the intensities of the gamma rays from ^{240}Pu are weak and often obscured by gamma rays from other isotopes. Plutonium-242 does not have a useful gamma ray, but its isotopic abundance is small for low-burnup plutonium. For high-burnup fuels such as LWR spent fuels, it can be estimated from correlations of other plutonium isotopes or inferred from results of an independent measurement of elemental plutonium concentration combined with gamma-ray spectrometry data for the other isotopes.

Samples of controlled and constant form and known chemical composition are the easiest for isotopic analysis. For example, if we prepare "thin" samples of low-concentration solutions in precision vials, we can obtain quantitative abundances of individual plutonium isotopes by comparing gamma-ray peak areas from standard solutions and applying self-attentuation corrections.

We have also developed a relatively simple method for the isotopic analysis of plutonium in samples of arbitrary shape and composition. The procedure requires no detailed peak fitting; thus it is fast, and computer cost and speed requirements are minimal. This method can be programmed into portable multiple-channel analyzers for use in field inspections. We measure areas under the peaks of closely spaced gamma-ray lines in the region from 120 to 414 keV to determine the following isotopic ratios.

^{238}Pu/^{241}Pu	152.7 keV/148.6 keV
^{239}Pu/^{241}Pu	203.5 keV/208.0 keV
	or 345 keV/332.3 keV
^{240}Pu/^{241}Pu	160.3 keV/164.6 keV
^{241}Pu/^{241}Pu	332.3 keV/335.4 keV
^{242}Am/^{239}Pu	125.0 keV/129.3 keV

In some energy regions, the measured lines are not clean peaks; that is, they must be corrected for contributions from neighboring lines of other isotopes. For example, the measured ^{240}Pu peak at 160.3 keV includes contributions from the ^{241}Pu line at 160.0 keV and from the ^{239}Pu line at 160.2 keV. By measuring the clean peak at 164.6 keV from the ^{241}Pu daughter ^{237}U, using the known intensity branching ratios for the 160.0- and 164.6-keV lines, and applying small corrections for changes in relative efficiencies, we can determine the 160.0-keV contribution and subtract it from the measured peak; similarly the ^{239}Pu peak at 161.5 keV is used to obtain the ^{239}Pu contribution to the 160-keV complex. The corrected peak is then divided by the ^{241}Pu (^{237}U daughter) peak at 164.6 keV to give the ^{240}Pu/^{241}Pu isotopic ratio.

For each sample, the relative detection efficiency is derived empirically from the areas of isolated, clean peaks of ^{239}Pu and ^{241}Pu in different regions of the spectrum, adjusted for their respective gamma-branching intensities. The isotopic ratios are then combined to yield the actual plutonium mass fractions using the constraint that their sum is unity. Results can be obtained by this method in 1 hour and, in some cases, with accuracies comparable to those obtained from mass spectrometry.

Active Gamma-Ray Assay

We are adapting two active gamma-ray assay methods for measuring fissionable materials in plant environments: x-ray edge densitometry and x-ray fluorescence. These methods com-

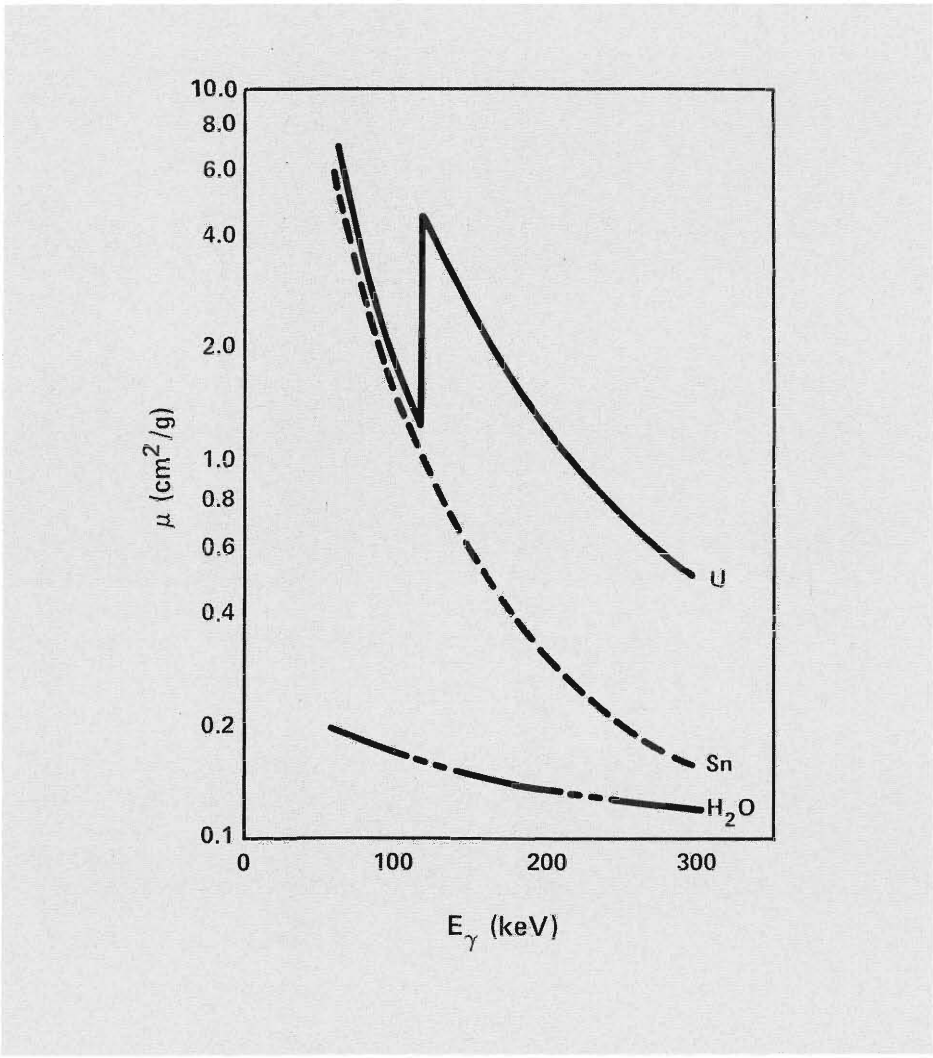


Fig. 11. Gamma-ray mass attenuation coefficient versus energy. The sharp K-edge discontinuity in the coefficient of uranium is contrasted with the monotonically varying coefficients of lower-Z materials.

TABLE II

ABSORPTION EDGES AND Δμ VALUES FOR URANIUM AND PLUTONIUM

Absorption Edge	Element	Edge Energy (keV)	Δμ (cm²/g)
L_{III}	U	17.17	56.6
	Pu	18.05	51.9
K	U	115.6	3.65
	Pu	121.76	3.39

plement passive gamma-ray assays in that they measure total elemental content rather than specific isotopes. The sample is bombarded with an external beam of photons or particles at energies tailored to induce specific atomic interactions characteristic of the element, such as removal of an electron from an inner atomic shell. Traditionally, analytical chemistry laboratories have implemented x-ray edge densitometry and x-ray fluorescence with x-ray generators and high-resolution wavelength dispersive techniques. Our adaptation uses high-resolution gamma-ray detectors to measure uranium and plutonium concentrations in solutions. The measurements can be made either in the process line itself (in-line) or by removing a sample and making the measurement alongside the process line (at-line). These techniques can be very useful in reprocessing plants to assay low-concentration solutions containing both uranium and plutonium.

We will limit this discussion to x-ray edge densitometry. The technique utilizes the fact that gamma rays with energies just above the threshold for removing an electron from the K or L_{III} atomic shells of uranium and plutonium are absorbed with much greater probability than lower energy gamma rays. These thresholds create jumps in the mass attenuation coefficient, known as K and L_{III} edges. By bombarding a sample with gamma rays just above and below the K or L_{III} edges, and measuring the difference in transmissions, we can measure the amount of plutonium or uranium in the sample.

The K edge for uranium is shown in Fig. 11, together with the smoothly varying mass attenuation coefficients of lower-Z elements in the same energy region. Table II presents the positions of the energies of the K and L_{III} edges of uranium and plutonium and the magnitudes of the corresponding mass attenuation discontinuities, Δμ.

The basic experimental arrangement for densitometry is shown in Fig. 12. Narrow-beam collimators eliminate the detection of multiple-scattered gamma rays and thereby minimize matrix effects. The transmission source is either an x-ray generator or appropriate radioactive isotopes. X-ray generators have the advantages of variable intensity and a continuous bremsstrahlung spectrum that can be tailored to cover the absorption edge, but radioactive sources are simpler and less expensive. High-resolution germanium detectors normally are used for K-edge measurements, whereas Se(Li) and germanium detectors must be used for L-edge measurements.

The transmissions T_1 and T_2 just below and above the absorption edge are related to the total concentration ρ of the element of interest by the relation

$$T_i = T_{m_i} e^{-\mu_i \rho X} ,$$

where

μ_i = μ_1 or μ_2, the mass attenuation coefficient of the element below and above its edges, respectively;
T_{m_i} = corresponding transmissions of the matrix (all other materials); and
X = sample thickness.

If the energies of the transmission gamma rays very closely bracket the edge, then $T_{m_1} \cong T_{m_2}$, and the concentration of the subject element is given by

$$\rho = \frac{-\ell n T_2/T_1}{\Delta\mu X} \; ; \; \Delta\mu = \mu_2 - \mu_1 .$$

Corrections for effects of matrix materials may be applied when the transmission gamma rays are not so closely spaced.

X-ray edge densitometry measurements for multiple simultaneous elemental determinations are shown in Fig. 13.

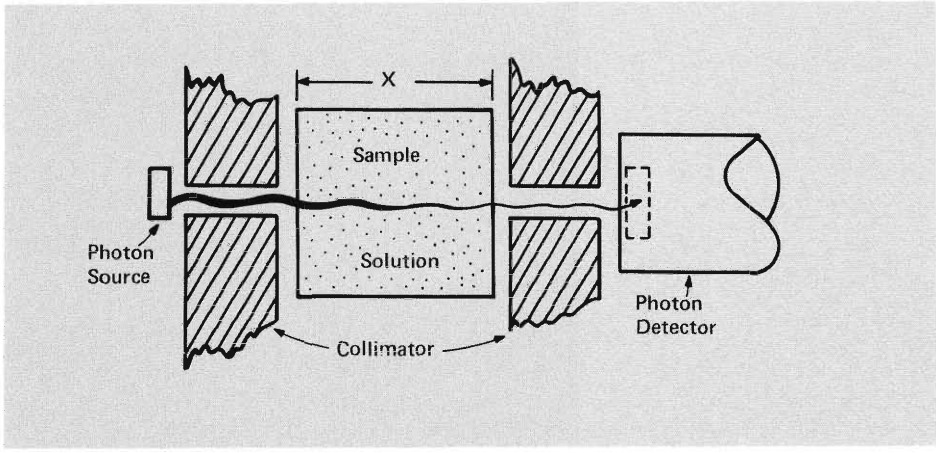


Fig. 12. Typical experimental setup for x-ray absorption-edge measurement of uranium and plutonium solution concentrations. The gamma-ray source is collimated, and the detector measures the intensity of gamma rays transmitted through the sample (thickness X) at energies above and below the characteristic x-ray absorption edges of the elements of interest.

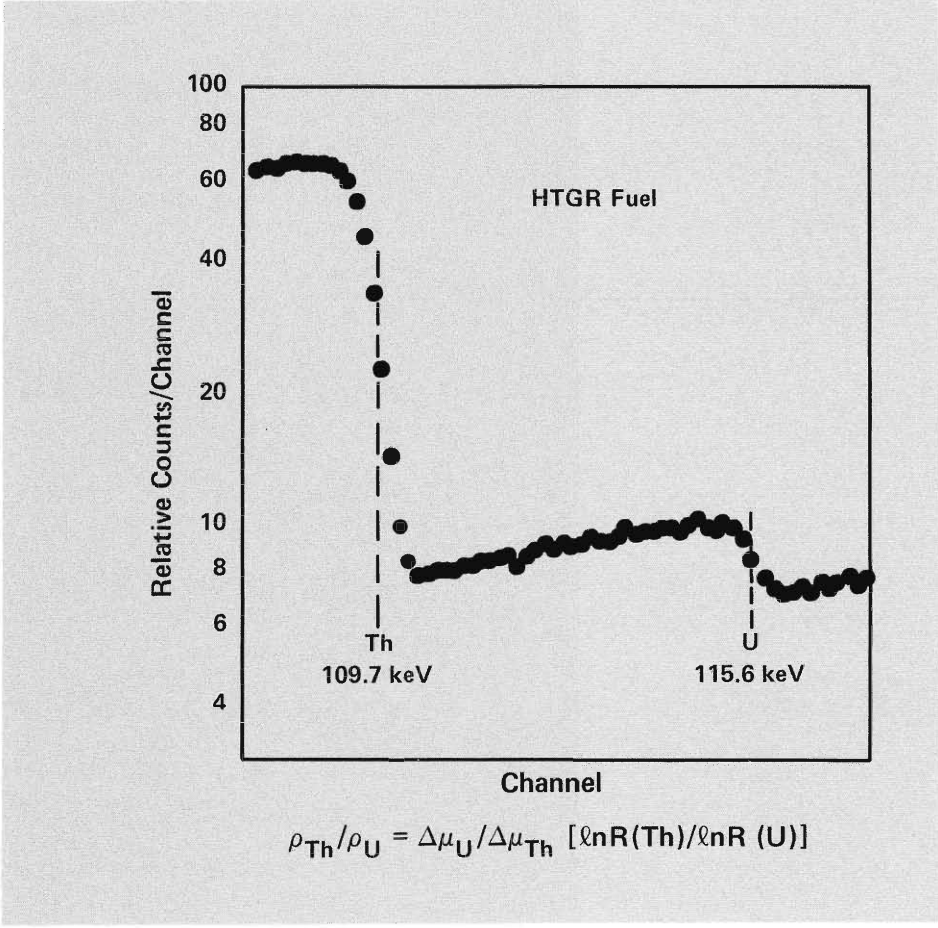


Fig. 13. Spectrum of x rays transmitted through a sample of high-temperature gas-cooled reactor fuel. The K edge of thorium at 109.7 keV and uranium at 115.6 keV are shown. Elemental concentrations are derived from the observed discontinuities at those edges.

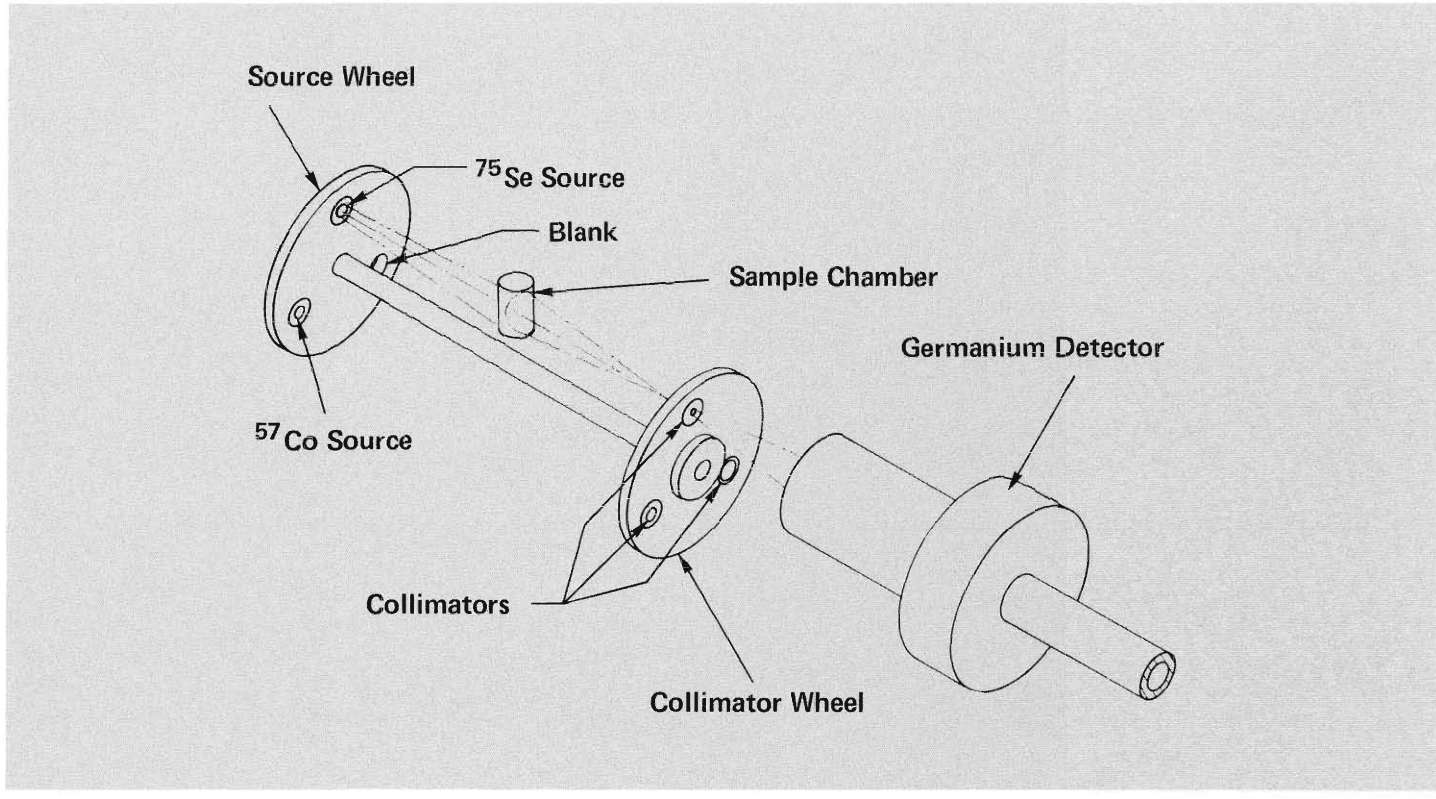


Fig. 14. Conceptual view of measurement configuration of the Tokai-Mura densitometer. The plutonium solution sample is shown between the source (^{75}Se, in this case) and the collimator in the detector line of sight. Sources and collimators are mounted on separate wheels driven synchronously.

The x-ray spectrum is transmitted through a sample of high temperature gas-cooled reactor (HTGR) fuel, which contains thorium and uranium in a graphite matrix. The discontinuities produced by the K-absorption edges of both uranium and thorium are clearly evident, allowing accurate determination of the respective densities of thorium and uranium, in this case in proportion 4:1.

To resolve low-concentration solutions of uranium and plutonium, we must use the L_{III} edge with its larger $\Delta\mu$ values rather than the K edge. The L_{III} edge is useful for solution concentrations ranging from 2 to 100 g/L. However, an x-ray generator is required for the transmission source because no adequate radioactive sources are available. We have developed an x-ray generator-based absorption-edge densitometer for the simultaneous measurement of uranium and plutonium in solutions containing both elements. Some future reprocessing and nitrate-to-oxide conversion plants will coprocess uranium and plutonium in such solutions. We have conducted a preliminary test of this instrument, operating in-line, at the Savannah River Laboratory coprocessing demonstration facility, as well as at LASL. The instrument performed well for mixed solutions with plutonium concentrations between 2 and 10 g/L and uranium concentrations 4 to 10 times higher and is thus a candidate for use in advanced, near-real-time accounting systems for fuel reprocessing and conversion facilities.

For K-edge measurements of higher concentration plutonium solutions, we can use two discrete gamma-ray transmission sources, the 121.1- and 122.2-keV gamma rays from ^{75}Se and ^{57}Co, respectively. These gamma-ray energies very closely bracket the 121.8-keV electron-binding energy of the K shell of plutonium.

We have recently developed such a densitometer to measure 150-350 g/L plutonium product solutions of the spent-fuel reprocessing in Tokai-mura, Japan. The instrument was installed and calibrated in this facility in the latter part of 1979 and is being evaluated for cooperative use by the facility operator and the IAEA for rapid off-line plutonium analysis. The Tokai densitometer rotates transmission sources in and out of position (Fig. 14) to measure the x-ray edge for plutonium concentration. This instrument also measures isotopic abundances by passive gamma-ray assay. Results obtained with the Tokai densitometer indicate that it will measure in times less than 1 hour plutonium concentrations to an accuracy of $\pm 0.3\%$ (1 standard deviation) or better, and isotopic abundances in the accuracy range 0.3-1.5%, depending on the isotope and its abundance.

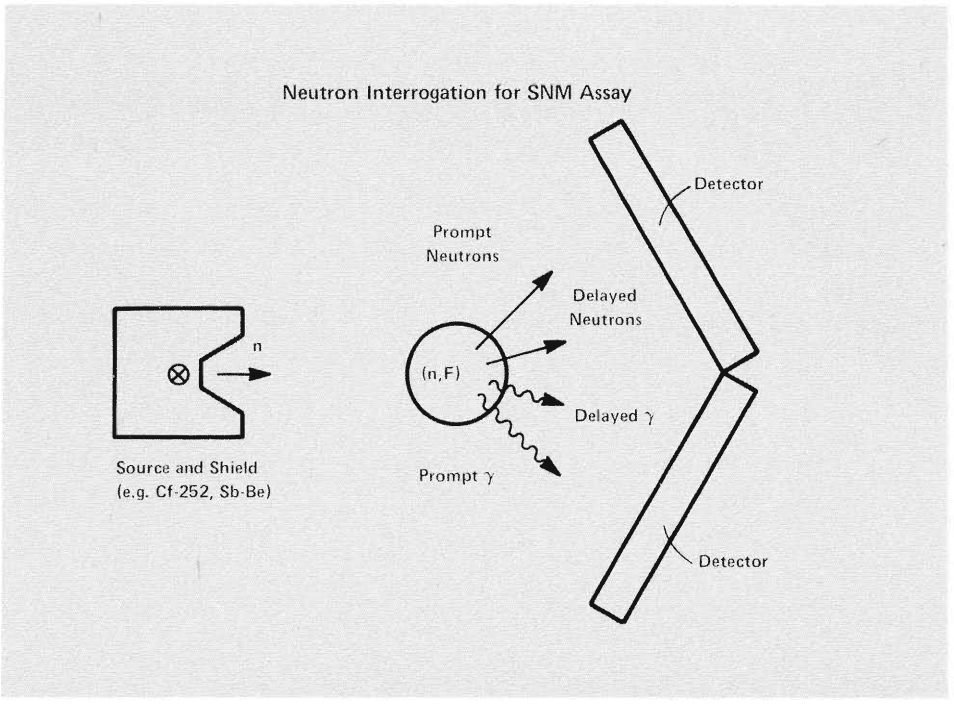


Fig. 15. The basic components of an active neutron assay system showing the interrogation source, the sample, and the detectors.

Passive Neutron Assay

Passive neutron assays detect the neutrons emitted when an isotope undergoes spontaneous fission. This is perhaps the easiest assay to perform. It is useful for measuring bulk materials because neutrons penetrate much farther than gamma rays. In practice, ^{240}Pu and ^{238}U are the only isotopes having sufficient spontaneous fission rates and isotopic abundances for passive neutron assay.

The assay is complicated by the fact that alpha particles from radioactive decays of uranium and plutonium interact with light elements in the matrix materials to produce additional neutrons. For example, if PuF_4 is present in the sample, neutrons will be produced through the reaction $^{19}F + \alpha \rightarrow {}^{22}Na + n$. However, each (α,n) reaction produces only one neutron in contrast to a spontaneous fission that produces a group of two or three. Consequently, we can differentiate spontaneous fission neutrons from the others by neutron coincidence counting. We have developed special time-correlation counting circuitry, called the shift register, to count coincidence neutrons and thus the spontaneous fission rate.

Neutron coincidence counting of ^{240}Pu spontaneous fission is the simplest method for the assay of plutonium provided the plutonium isotopic composition is known ahead of time. For high-burnup plutonium reactor fuels, a coincidence count may also contain a significant contribution from ^{242}Pu spontaneous fission. Since the spontaneous fission rate for ^{240}Pu is low, approximately 460 fissions/s per g ^{240}Pu, and the ^{240}Pu isotopic abundance is typically in the range 8-20%, a high-efficiency detection system is essential for rapid, quantitative assay. For uranium fuels, ^{238}U can be assayed by counting its spontaneous fissions, but the specific rate is much lower (approximately 7×10^{-3} fissions/s per g ^{238}U) and kilogram quantities are required. We have developed cylindrical detectors (with a central sample cavity) that have efficiencies as large as 20% for coincidence detection of fission events. These detectors are made of ^3He or BF_3 proportional counters and polyethylene. The sensitivity of the highest efficiency units is about 1 mg ^{240}Pu. Organic scintillation detectors have also been used for coincidence detection of spontaneous fission.

Active Neutron Interrogation

When the gamma-ray attenuation of materials is too high for passive assay, and the spontaneous fission rate is too low for neutron coincidence counting, we can interrogate the sample with highly penetrating beams of neutrons. The bombarding neutrons induce fissions in the sample and various gamma-ray and neutron signatures of the neutron-fission reactions are detected. During the past few years, active neutron interrogation has been used extensively to measure uranium and plutonium fuel rods, unirradiated fuel assemblies, and high-enriched uranium scrap. Either accelerator or radioisotopic neutrons could be used. Although accelerator neutron sources offer large yields and source modulation flexibility (that is, pulsing), they usually require a substantial amount of initial investment and technical support. On the other hand, radioactive neutron sources are reliable, simple, and less costly and are well suited for in-plant measurements and quality control of reactor fuel components as well as for some categories of scrap-material and process-stream measurements.

The basic components of an active neutron assay system are the neutron source, the sample, and the detectors to count the induced activities (Fig. 15). The neutron source is normally surrounded by materials to moderate or slow down the neutrons and tailor their energy for interrogation. These materials also are needed for personnel shielding. The fissions induced by the irradiation emit prompt and delayed neutrons as well as prompt and delayed gamma rays. One or more of these emitted signatures can be used for the assay depending on complexity of the measurement problem and the detection method.

Most fissionable material contains a mixture of both fissile and fertile isotopes. The fertile isotopes (for example, ^{238}U and ^{232}Th) have a threshold energy (about 1 MeV) for induced fis-

TABLE III
A NEUTRON IRRADIATION TECHNIQUE

Moderator Configuration[a] (Thickness in cm)	Total Leakage (%)	Median Energy (MeV)	Fraction of Neutrons With E < ^{238}U Threshold (%)	Fission Ratios[b] (^{235}U/^{238}U) Measured	Calculated
Pb: 7:5 (Cd)	146	1.76	39	003.19	003.10
W: 7:5 (Cd)	135	0.47	78	005.68	006.53
W/C: 7:5/7:5 (Cd)	122	0.19	79	015.3	013.3
W/C/CH_2: 7:5/7:5/2:5 (no Cd)	109	0.002	80	600.	788.

[a]The moderators and detectors were covered with 0.76-mm-thick cadmium where noted. DTF-IV calculations were used for the moderators with no cadmium; all others used the Monte Carlo code.

[b]Fission ratios are for equal weights of each fissionable isotope.

sion reactions, whereas fissile isotopes (for example, ^{235}U, ^{233}U, and ^{239}Pu) fission for all neutron energies. Active neutron interrogation techniques use subthreshold and superthreshold irradiation to separate the fissile and fertile components in a sample. The D-T, 14-MeV neutron generators are a convenient source of these interrogation neutrons, but the neutron energy is so high that both fissile and fertile isotopes undergo fission. To separate these components, we have developed neutron tailoring techniques to give both superthreshold and subthreshold irradiations from the same 14-MeV source. We have used Monte Carlo and DTF-IV neutron transport calculations to obtain maximum neutron leakage and desired neutron energy characteristics from various moderator assemblies. For example, for a 14-MeV neutron source, we have designed moderating assemblies that consist of concentric spherical shells of different materials including tungsten, graphite, and polyethylene. The large cross sections for the (n,2n) and (n,n′) reactions (incoming neutrons, outgoing neutrons) in tungsten reduce the energy of the 14-MeV source neutrons to a median energy of approximately 500 keV, and elastic scattering of neutrons in the graphite further reduces the average neutron energy to the low-keV region. Table III lists typical moderator configurations, where the notation W/C/CH_2: 7.5/7.5/2.5 indicates a 7.5-cm-thick core of tungsten surrounded by a 7.5-cm-thick shell of graphite surrounded by a 2.5-cm-thick shell of polyethylene. The total neutron leakage from the moderators exceeds 100% because of the large (n,2n) cross section in the heavy isotopes. Table III shows that the median neutron energy can be lowered from 14 MeV to a few keV using moderators with radii less than 18 cm. The tailoring assemblies given in Table III result in ^{235}U/^{238}U fission ratios from 3-600, a range that is very adequate for the separation of the fissile and fertile components in assay applications.

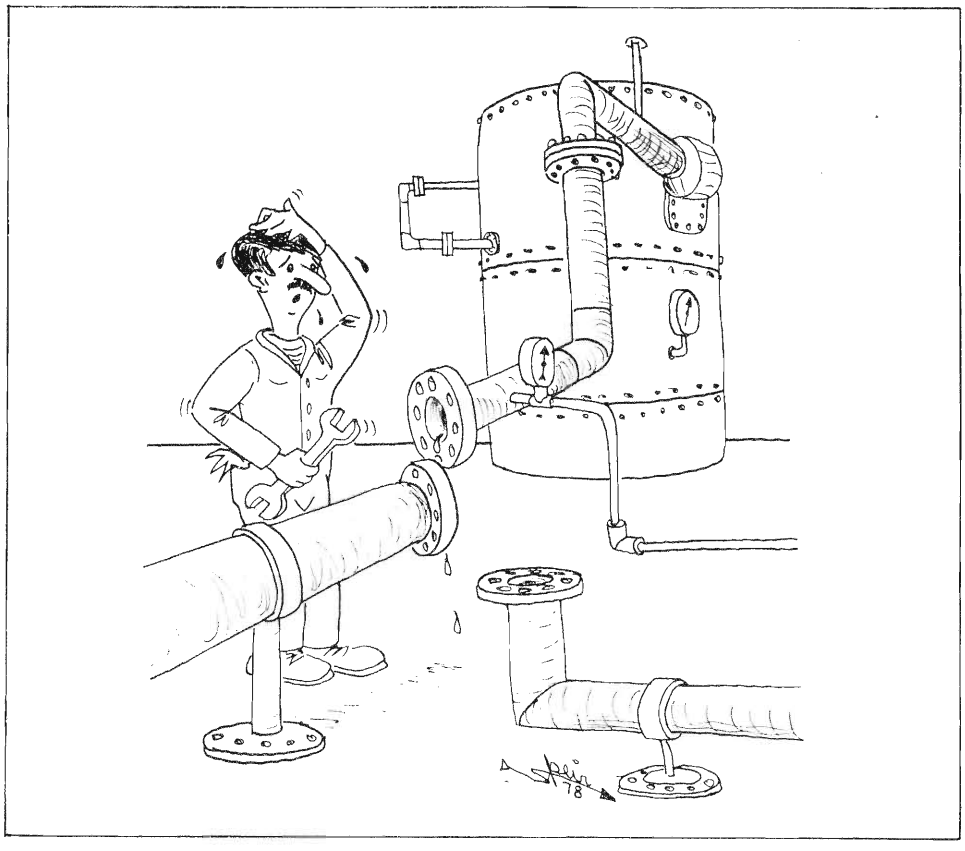


TABLE IV
RADIOACTIVE SOURCES FOR ACTIVE ASSAY APPLICATIONS

	Prompt n and γ		Delayed n and γ
Neutron Source	**Integral**	**Coincidence**	**Integral**
Sb-Be; 25 keV	n		
Ra-Be; 200 keV	n		
^{238}Pu-Li; 400 keV	n		
Am-Li; 400 keV		n and γ	
^{252}Cf; moderated		n and γ	n and γ
^{252}Cf; thermalized	n	n and γ	n and γ

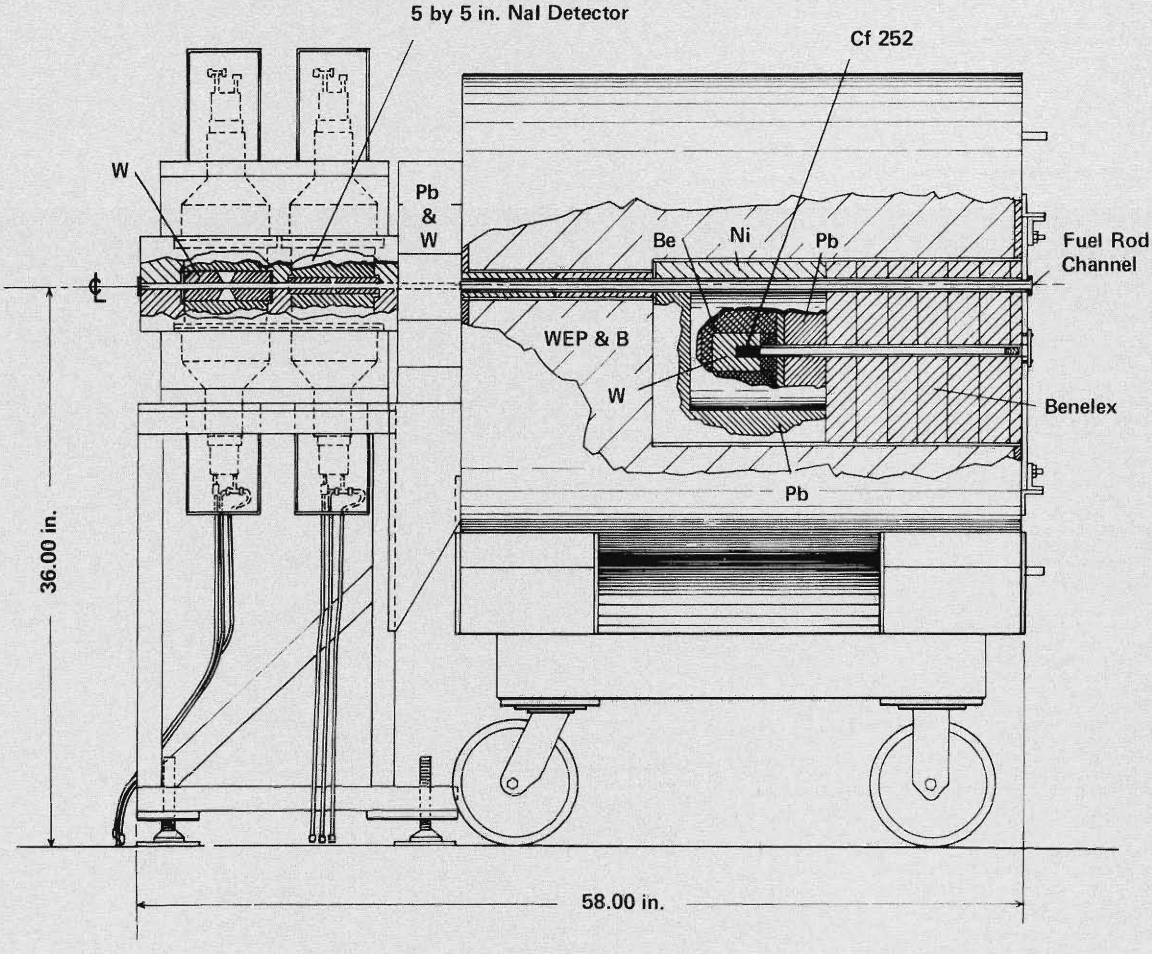


Fig. 16. Schematic of the ^{252}Cf fast-neutron assay system for the fast breeder reactor fuel rods. The delayed gamma rays induced by the fast-neutron irradiation are subsequently counted with the two NaI detectors, which also measure the passive gamma rays to detemine pellet-to-pellet uniformity.

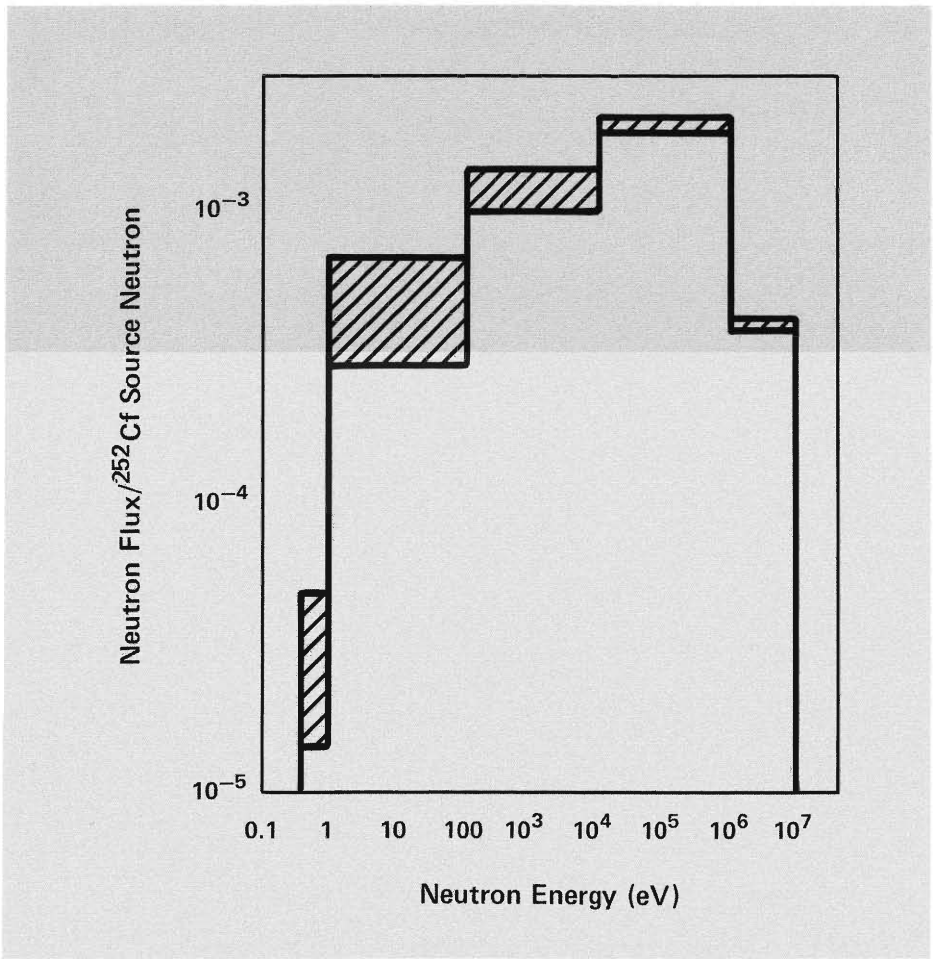


Fig. 17. Calculated neutron spectrum from a ^{252}Cf source for the system shown in Fig. 16 with (cross-hatched area) and without the cylindrical nickel fast-flux trap.

For most safeguards applications employing active neutron assay, radioactive sources have been preferred because of their simplicity and reliability.

To apply radioactive sources to a given measurement problem, three primary variables should be optimized: (1) the type of neutron source, (2) the moderator and shield for the source, and (3) the detector to count the induced neutrons and/or gamma rays. Radioactive neutron sources under investigation at LASL include ^{124}Sb-Be(γ,n), ^{88}Y-Be(γ,n), ^{226}Ra-Be(γ,n), and Am-Li(γ,n) (which have neutron energies primarily below the fission thresholds of ^{238}U and ^{232}Th), and ^{252}Cf (which emits higher energy neutrons).

Table IV gives a summary of radioactive sources and corresponding detection methods that have been used by LASL for assay applications. These applications range from portable field instrumentation used by inspectors to large in-plant installations used for waste and spent-fuel assay.

Fuel Rod Scanners

A high-performance instrument that incorporates several of the assay signatures previously described is the fuel rod scanner for the Fast Flux Test Facility (FFTF) at Hanford, Washington, a low-power reactor used to test designs for the Breeder Reactor Program. The design of the scanner required extensive use of Monte Carlo computer calculations to determine an optimum combination of materials, geometry, and detectors.

This hybrid assay system uses active neutron interrogation to determine total fissile content and passive gamma-ray spectroscopy to measure selected plutonium isotopes. The system measures the plutonium fissile content in a fuel rod to better than 0.5% accuracy and verifies that the plutonium content in individual fuel pellets within each rod is uniform from pellet to pellet. The fuel rods contain 70-80% natural uranium and 20-30% plutonium. The rods have an active length of 914 mm and a diameter of about 5 mm. Many thousands of these rods are used in the reactor core, and tight accountability of the plutonium is required for nuclear material safeguards.

A schematic of the system (Fig. 16) shows the 600-μg ^{252}Cf neutron source used for the interrogation, the moderator, the radiation shield, and the NaI detectors used to count the gamma rays. The neutron moderator is designed to give a fast-neutron irradiation (the irradiation channel is lined with cadmium) and a high fissile/fertile fission ratio (400/1). The fissile/fertile fission ratio corresponds to

$$\frac{\int \sigma^f_{239}(E)\,\phi(E)\,dE}{\int \sigma^f_{238}(E)\,\phi(E)\,dE}\,,\quad E > 0.4\ \text{eV}\,,$$

where σ^f_{239} and σ^f_{238} are the ^{239}Pu and ^{238}U fission cross sections, respectively, and $\phi(E)$ is the energy-dependent neutron flux in the irradiation channel.

Neutron transport calculations using both S_n and Monte Carlo techniques were used in the design of the ^{252}Cf source moderator. The calculations led to the design of the cylindrical moderator assembly shown in Fig. 16 with a tungsten core surrounded by a shell of beryllium followed by nickel and lead for fast-neutron reflection. The calculated neutron spectrum from a ^{252}Cf source for this system is shown in Fig. 17 with (cross-hatched area) and without the cylindrical nickel fast-flux trap. The presence of the nickel reflector increased the fission rate by approximately 70% over the lead reflector. This moderator resulted in a ^{239}Pu/^{238}U fission ratio of approximately 400/1 for irradiation neutrons above the cadmium cutoff energy (0.4 eV). It is necessary to keep this ratio high to avoid a significant contribution from ^{238}U fission, which is not of safeguards interest.

During operation, up to 30 fuel rods are placed in the loading magazine of the

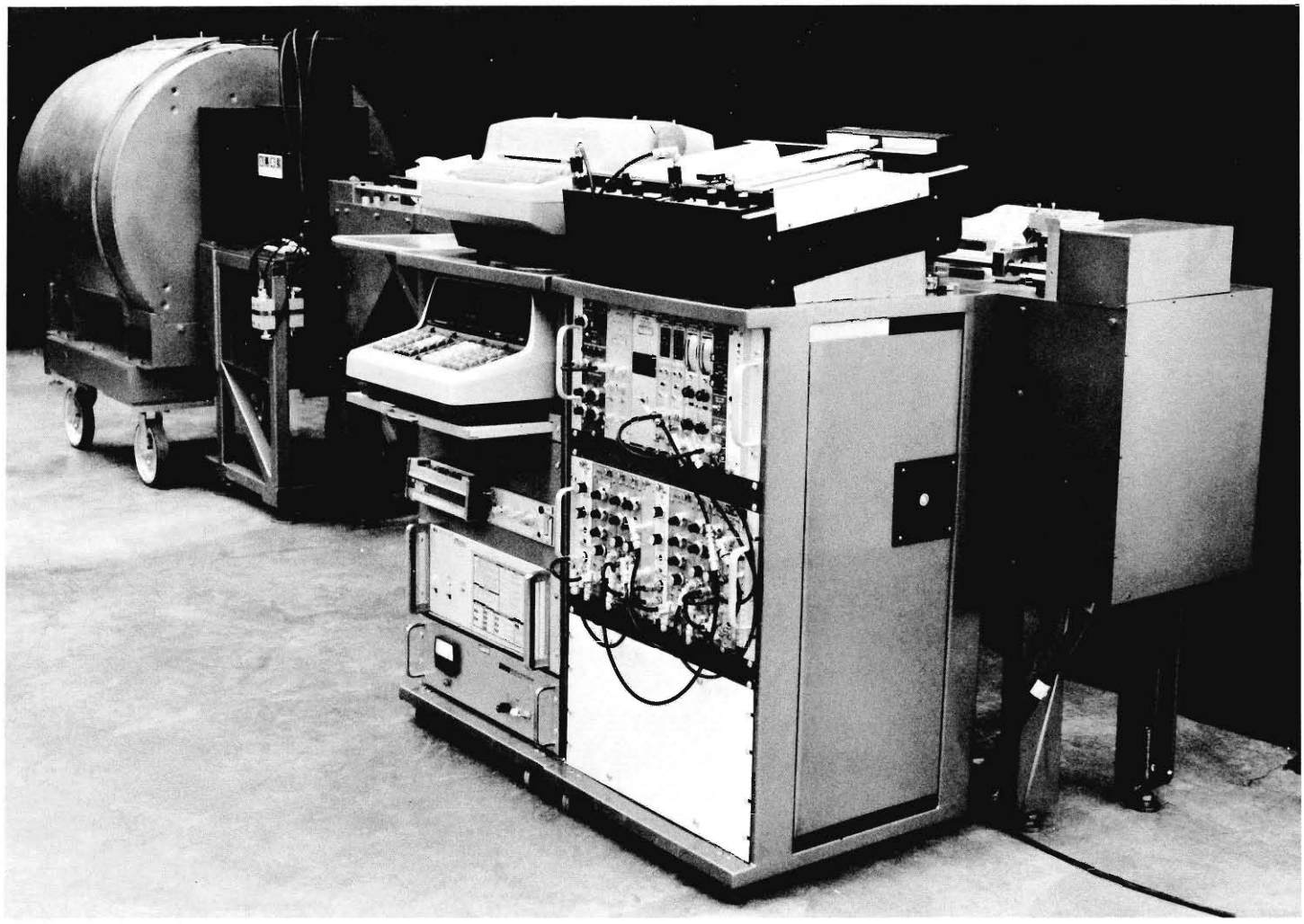


Fig. 18. Fast-neutron ^{252}Cf assay system for fast breeder reactor fuel rods. The system includes a 600-μg ^{252}Cf source and shield, two 5- by 5-in. NaI detectors to count the delayed gamma rays, automated fuel-rod handling, and a data-reduction system.

fuel-rod translator. The automated translator picks up the rod to be assayed and moves it through the NaI crystals to take a background count of the unirradiated rod and then into the ^{252}Cf assembly for irradiation with neutrons. The direction of travel is then reversed and the rod is withdrawn through the NaI detectors, which measure the delayed fission gamma rays for the total fissile assay and the passive gamma rays for the pellet-to-pellet uniformity. The two 5-in.-long by 5-in.-diam NaI detectors count the high-energy delayed gamma rays (>1200 keV) to determine the total fissile measurement because of their higher penetration through the rod. Simultaneously, the same NaI detectors determine pellet-to-pellet uniformity by measuring the much more intense lower energy passive gamma rays from the fuel. Each of the two NaI detectors examines a different low-energy window on a pellet-to-pellet basis, using a tungsten sleeve with a narrow collimation slit. One detector counts 60-keV gamma rays, which are mainly from ^{241}Am, and thus serves as a batch monitor because the ^{241}Am content of the fuel is time dependent. The second detector counts primarily plutonium gamma rays in the range of 100 to 500 keV. This second window provides a qualitative measure of the fissile plutonium in the rod. After the irradiation and scan of the fuel column, the rod is unloaded in the tray directly below the loading magazine and the cycle is repeated for the next rod.

The complete assay system, which includes automated fuel rod handling and computer-based data reduction, is shown in Fig. 18. This system was installed in 1973 at the Hanford Engineering Development Laboratory (HEDL) Plutonium Facility and has been used to measure the plutonium fissile content (better than 0.5% accuracy) and uniformity of over 70 000 FFTF fuel pins, about 40 000 of which are now being used in the reactor.

The ^{252}Cf-Based Assay System for the FAST Facility

We have designed a neutron interrogation assay system to measure waste solids from reprocessing and spent-fuel packages at the Fluorinel and Storage (FAST) Facility, a new addition to the Idaho Chemical Processing Plant used to reprocess spent fuels from the national defense program. Measurements of the canisters of solid waste will be performed for ^{235}U process control and accountability of the waste solids. The spent-fuel packages will be assayed to provide assurance that the ^{235}U content is below 10.5 kg, the criticality limit

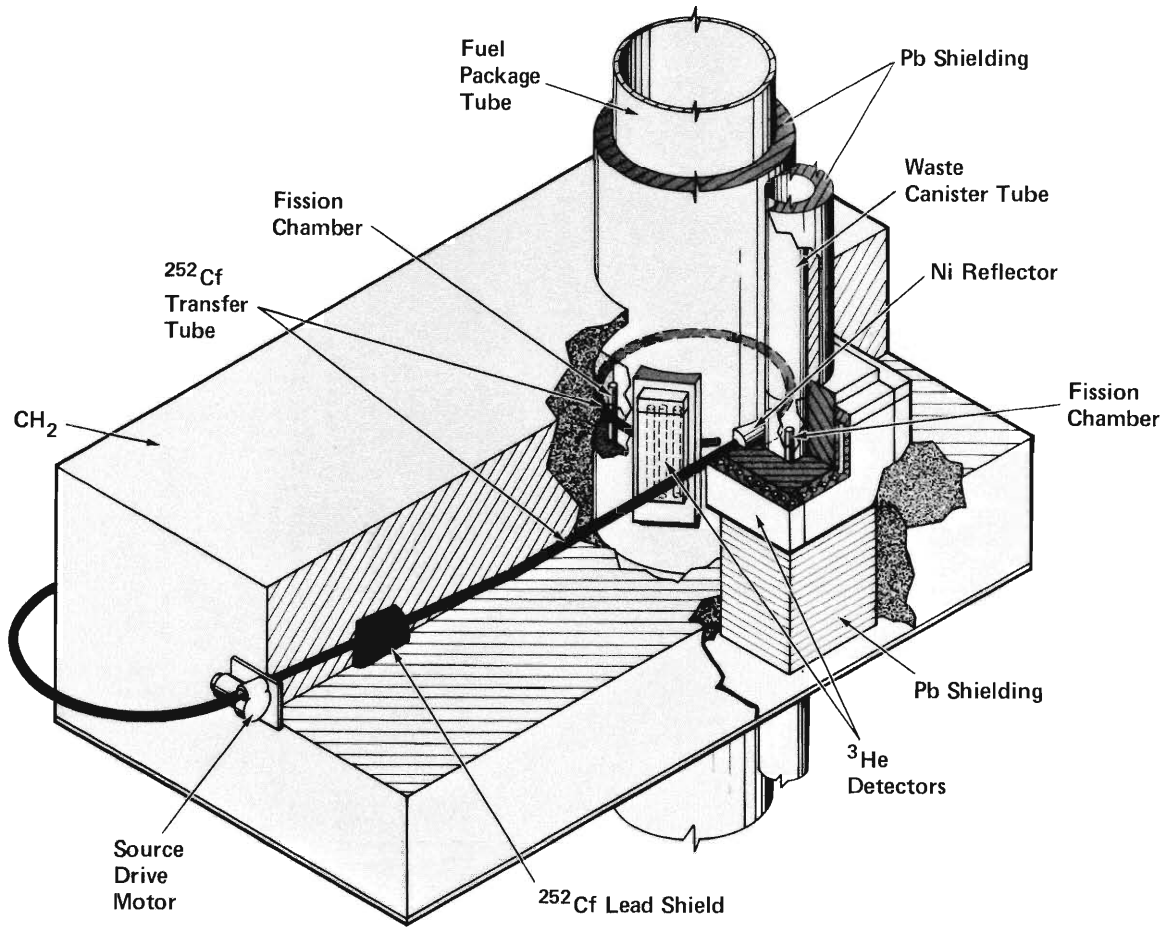


Fig. 19. The fast-neutron assay system for the Idaho Chemical Reprocessing FAST facility showing ^{252}Cf source transfer system, ^{3}He detectors, and the sample tubes for irradiated fuel elements and waste containers.

The assay system is based on the principle of the *shuffler*, which is a modulated ^{252}Cf source assay system that repetitively transfers the neutron source from the interrogation position to a shielded storage position while the delayed neutrons are counted. The assay system shown in Fig. 19 includes a source shield tank, a decoupling CH_2 shield, and an irradiation area. The assay sample is irradiated in a chamber surrounded by ^{3}He neutron detectors that are used to count delayed neutrons produced following each irradiation. We have evaluated the shuffler system for the assay of fuel rods, inventory samples, scrap and waste, uranium ore, irradiated fuel, and plutonium mixed-oxide fuel.

At the FAST Facility, the fissile contents vary from 0 to 400 g in the waste and from about 8 to 12 kg in the spent-fuel elements. Measurements must be obtained in the presence of large neutron (1.2×10^7 n/s) and gamma (50 000 R/h) backgrounds. The system employs fast-neutron interrogation of the sample using a large (5-mg) ^{252}Cf source to override the neutron backgrounds. Preliminary Monte Carlo calculations indicate that measurement precisions of better than $\pm 5\%$ ($2\text{-}\sigma$) are obtainable for waste loadings in excess of 200 g ^{235}U.

We are designing the assay instrument as an integral part of the FAST Facility with close cooperation from Exxon and Ralph M. Parsons Company (Architectural Engineer). The facility will house the assay instrument in a separate cubicle connected by through tubes to the fluorinel dissolution cell. Waste canisters and spent-fuel packages will be lowered by crane into the assay instrument through the tubes that penetrate the cell floor and extend below the assay system. The assay system will be remotely controlled and operated by a dedicated mini-computer system. Design of the instrument as an integral part of the facility, rather than as a retrofit, enables both facility operations and assay measurements to be better coordinated.

The Active Well Coincidence Counter (AWCC) for Portable Inspection Applications

An important safeguards area that has very different requirements from in-

Fig. 20. The Active Well Coincidence Counter including detector body, cart, and portable electronics for automated data collection and analyses.

plant instrumentation is field inspection using portable instrumentation. Equipment for this application must be lightweight, rugged, and simple to operate. A leading example of this instrumentation is the High-Level Neutron Coincidence Counter (HLNCC), which measures the effective ^{240}Pu content in bulk plutonium samples by coincidence counting the spontaneous fission neutrons from the plutonium.

To extend the same type of equipment to uranium assay applications, we use active neutron interrogation to induce the fission reactions because the spontaneous fission rate in ^{235}U is too small to be useful in the passive mode.

The basic principle of the AWCC is fast-neutron interrogation using a random neutron source (for example, AmLi) and counting the induced fission reactions using coincidence techniques to suppress the signal from the random interrogation source. The AWCC uses 42 ^{3}He detectors to count the induced fission neutrons. It also has two neutron sources of similar strength ($\sim 5 \times 10^4$ n/s), one in the lid and one in the bottom plug. The use of two neutron sources produces a rather uniform vertical response. Also, the CH_2 plugs act as neutron shields to reduce the background neutrons counted in the ^{3}He tubes from the AmLi sources. This shielding technique improves the induced signal-to-interrogation neutron background ratio by a factor of 10.

The AWCC has been designed to take advantage of the portable electronics package that was developed for the HLNCC. The electronics unit is interfaced directly to the HP-97 programmable calculator shown in Fig. 20. A microprocessor in the unit reads out the run time, total counts, real plus accidental counts, and accidental counts to the HP-97. The HP-97 reduces the data using the software package selected by the operator.

The unit is useful for measuring bulk UO_2 samples, high-enrichment uranium metals, LWR fuel pellets, and ^{233}U-Th fuel materials, which have very high gamma-ray backgrounds. By removing the AmLi source, the unit can measure ^{238}U and plutonium in the passive neutron coincidence mode. We recently have supplied the first AWCC system to the IAEA for field test and evaluation.

Status of Development and Implementation

Acceptable performance criteria for NDA techniques for safeguards, established well over a decade ago, include measurement times in the range from a few seconds to 1 hour and the following accuracies for the various material categories: 0.2-3.0% for uniform feed and product materials, 2-10% for recoverable scrap, and 5-30% for waste. We have achieved or exceeded these goals for many of the materials associated with the following fuel processes: uranium enrichment, uranium and plutonium fuel fabrication, and uranium and plutonium scrap recovery. The development of NDA methods and instruments has been greatest for measurements of scrap and waste, holdup, high-purity raw materials, and finished fuels—the material categories that were first identified as priority problems for NDA development.

Portable instruments and a number of stand-alone instruments for measuring containers of SNM were the first to be developed and tested in operating facilities. Some of these instruments then were adapted for on-line applications so that items or samples of plutonium material could be measured without being removed from the glove box containment.

Current research is focused on extending the range of applicability (to other compositions and concentrations) and improving accuracies of existing technologies, adapting techniques for in-

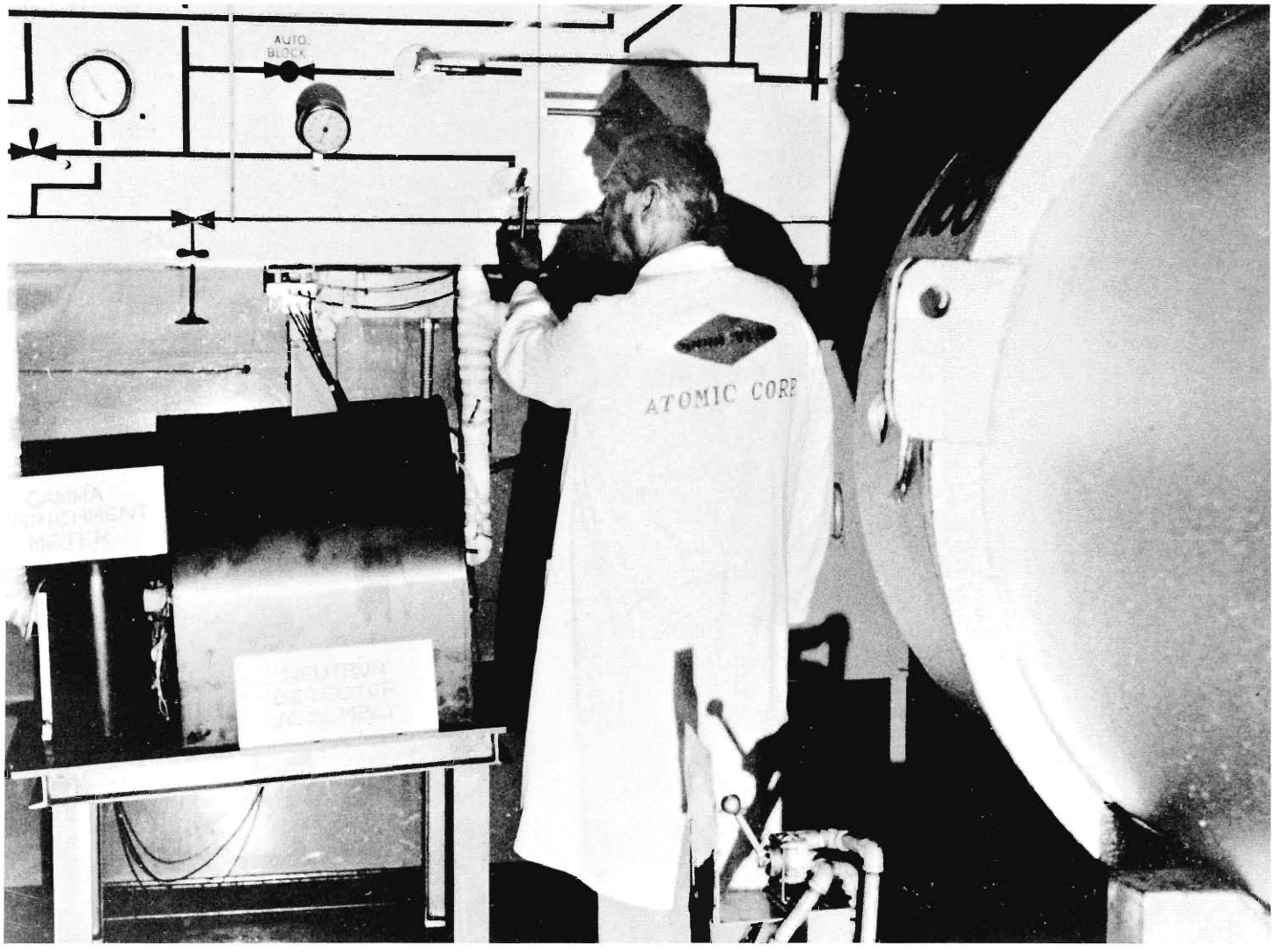


Fig. 21. In-line UF_6 enrichment monitor installation at the extended-range product-withdrawal station of the DOE/Goodyear Atomic gaseous diffusion plants. Passive gamma-ray and neutron instruments continuously measure ^{235}U and ^{234}U enrichments of liquid UF_6 to provide criticality safety and process controls.

line measurement and monitoring of bulk process materials, developing specialized instrumentation for inspectors and for containment and surveillance, developing methods to measure spent fuels and "hot" reprocessing materials, and finding techniques to measure advanced fuel materials such as mixed uranium-plutonium solids and solutions.

Ultimately, the best gauge of the success of these developments is the level of acceptance and implementation by the nuclear industry, regulatory agencies such as the US Nuclear Regulatory Commission, and for the international arena, the IAEA. In most operating nuclear facilities that process low-enriched uranium for use in commercial power reactors and high-enriched uranium and plutonium for defense programs and research reactors, the implementation of the NDA methods described here to complement or replace analytical chemistry measurements for safeguards and process control has been extremely gratifying. An example is the in-line UF_6 enrichment monitor at the DOE/Goodyear Atomic gaseous diffusion plant (Fig. 21), which serves principally for process control and criticality safety. Major exceptions are spent-fuel reprocessing plants, where the slowdown or postponement of reprocessing has in turn delayed the testing and implementation of promising measurement methods for the fission-product-contaminated

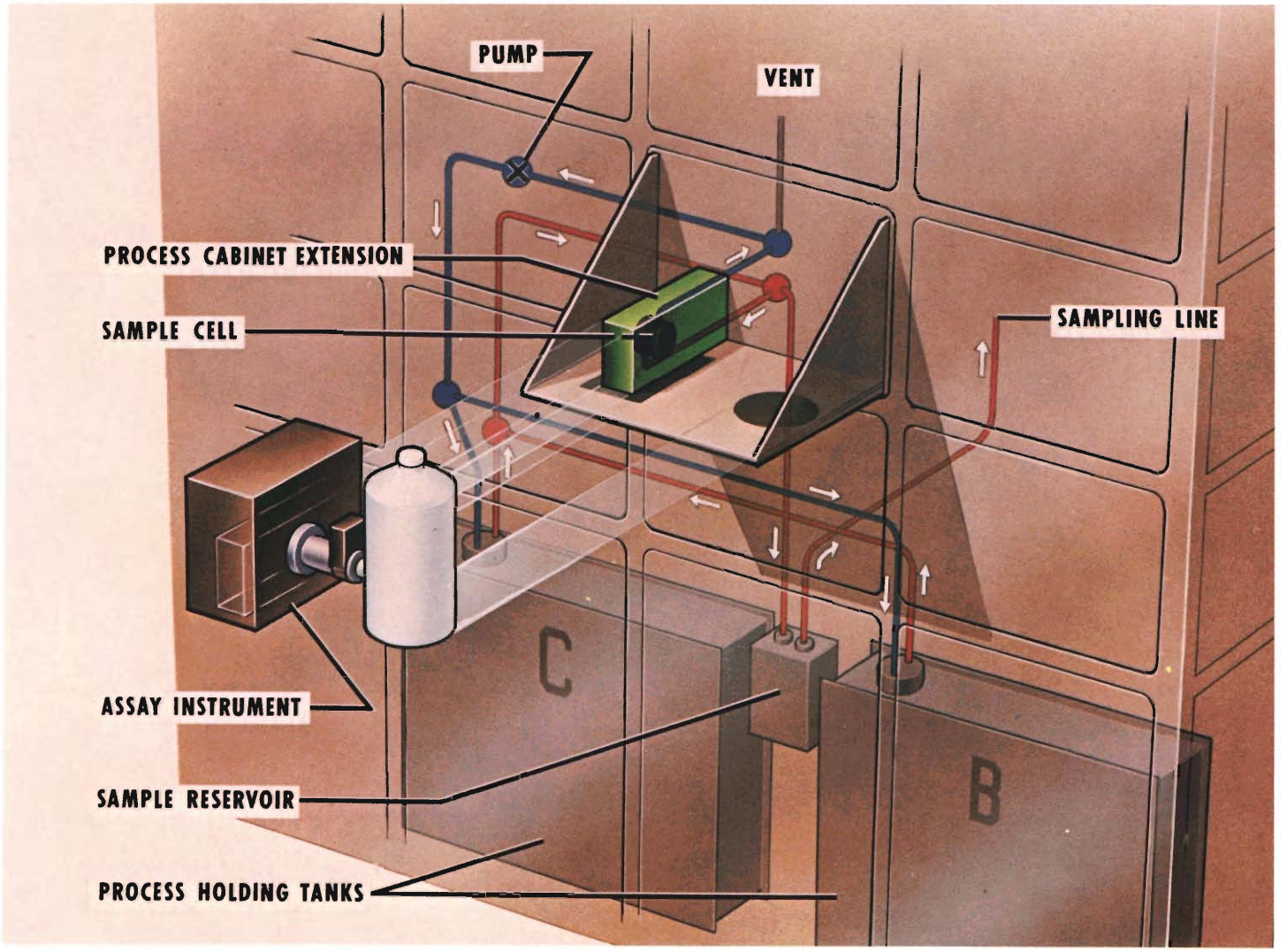


Fig. 22. The in-line absorption-edge densitometer for the Savannah River Reprocessing Plant. Plutonium product solution is pumped from holding tanks through a measurement cell, which is located inside an extension of the process line containment.

uranium and plutonium materials processed in these plants. A project is now under way to test an in-line K-edge densitometer (Fig. 22) for measuring plutonium concentration in the product solution line of the Savannah River spent-fuel reprocessing plant, which is operated for DOE national defense programs.

A typical nuclear facility now has an NDA counting room in which scrap and waste and samples from the process lines are measured for materials accounting, process control, and waste management. Portable NaI and germanium gamma-ray detectors and neutron counters are used routinely to measure holdup for inventories or investigate process anomalies. Fuel rod scanners are used in all light water reactor fuel fabrication facilities in the United States and most of those in Belgium, France, Germany, Italy, and Japan to quality-assure all fuel rods. Some facilities, such as the plutonium plants operated at LASL and Rocky Flats for defense programs and the General Electric/Wilmington light water reactor fuel fabrication plant, have installed near-real-time material accounting systems that use NDA instruments for measurement or verification of material movement throughout the plant.

We are equally gratified by the acceptance of NDA technology by regulatory agencies, both as a part of safeguards systems for plants and for independent

TABLE V
INSTRUMENTATION DEVELOPED BY LASL FOR IAEA APPLICATIONS

Instrument	Principle	Applications
SAM-II	Passive gamma-ray enrichment meter	UF_6 cylinders, UO_2 powder, fuel pellets and rods
Segmented gamma scanner	Transmission-corrected gamma-ray measurements	UO_2, PuO_2 scrap and waste
HLNCC	Passive neutron coincidence counting	Pu metal, PuO_2 powder, MOX fuel
AWCC	Active neutron interrogation and coincidence counting	U metal, UO_2 powder, U-Al alloy
Coincidence collar	Active neutron interrogation	PWR, BWR, and HWR fuel assemblies
K-edge densitometer	Active gamma-ray transmission	Pu and U solutions reprocessing plants
Reactor power monitor	Passive neutron	LWR reactor power monitor
Spent-fuel verification instruments	Cerenkov glow, passive gamma-ray, passive neutron	LWR and HWR spent fuel assemblies in storage pools

inspector verifications. Through the US program of technical assistance to the IAEA, we have developed a number of key NDA instruments and applications for international inspection of nuclear plants, and have provided inspectors with training, manuals, calibration, and implementation assistance. Table V lists instrumentation we have developed for IAEA applications. One technique helps to verify relative burnup and cooling time of spent reactor fuel assemblies to infer the resulting plutonium inventory (Figs. 23 and 24). E. Dowdy has pioneered a convenient method using the Cerenkov glow for confirming that the spent-fuel assemblies stored in an underwater array as shown in Fig. 23 are

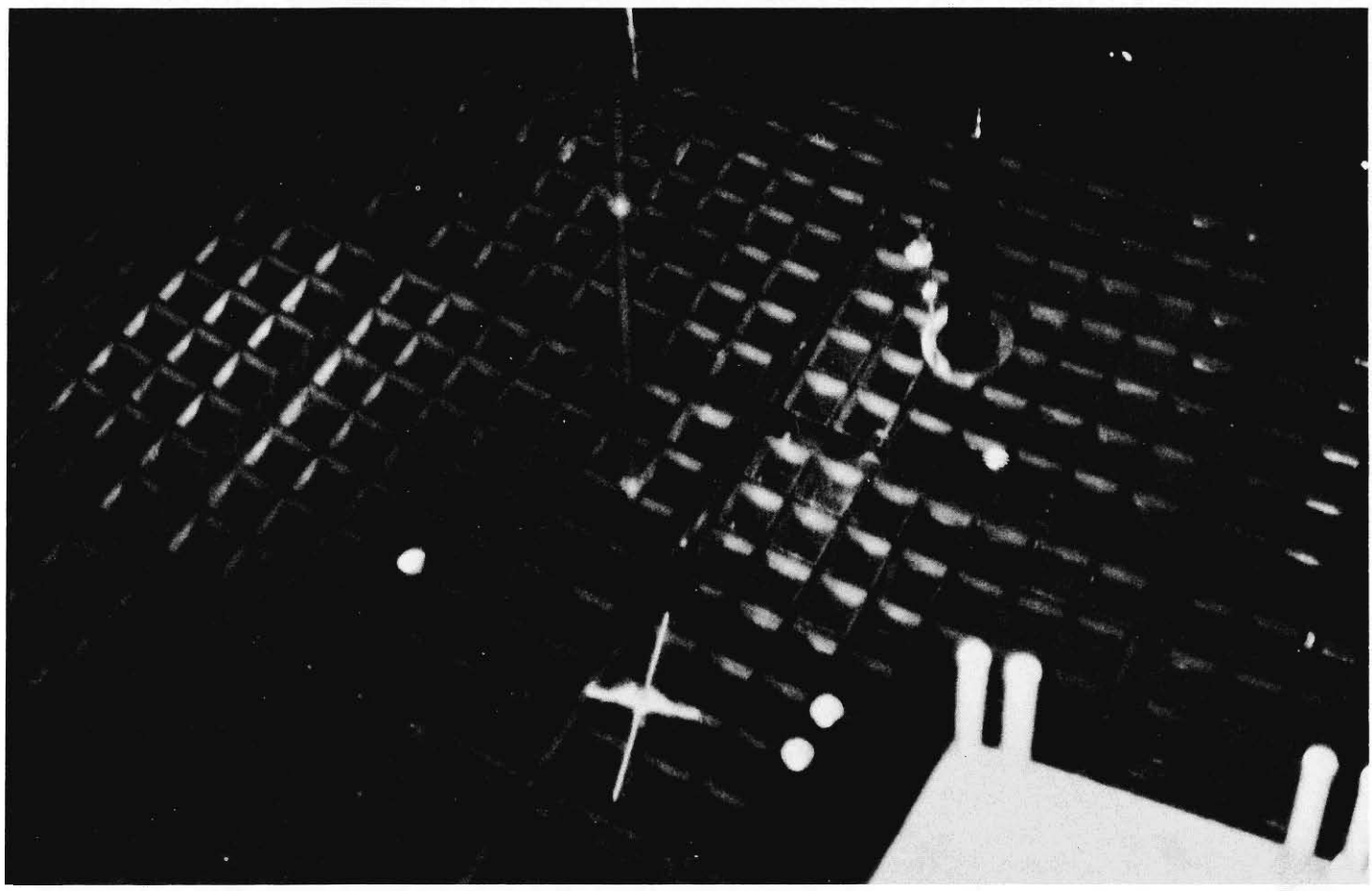


Fig. 23. A light water reactor fuel element storage array under about 10 m of water in a spent-fuel storage pool. One fuel element is raised about one-half its length through the ring ion and fission chamber (see Fig. 24) to measure the gamma-ray and neutron emissions.

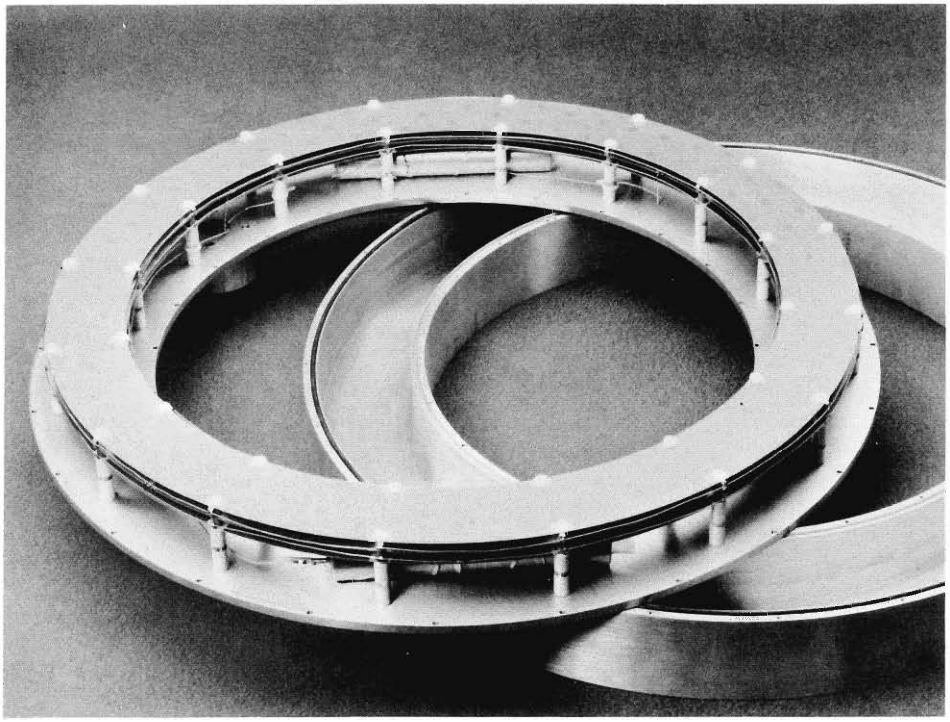


Fig. 24. Ring detector containing ion and fission chambers to measure intense gamma-ray and neutron emissions from spent-fuel assemblies to help verify burnup and cooling time in the fuel.

highly radioactive. Inspectors can turn off the overhead lights and measure the intensity of the Cerenkov glow, which is proportional to the activity level in the spent-fuel assembly. This approach is rapid and requires no instrumentation to be put in the contaminated water of the pool.

Challenges of the Future

While great progress has been made in filling the voids in safeguards measurement technology with NDA methods, the implementation of new technology has been confined largely to retrofitting existing facilities and conventional safeguards procedures. We have introduced high technology for measurements and accounting into plants that were designed both for manual operations and to rely on administrative controls and chemical assay for safeguards accounting. Implementation of NDA-based vehicle and personnel monitors has enhanced the containment and sur-

veillance elements of safeguards for existing facilities.

Heretofore, the development of instruments has been focused on the need to measure uranium and plutonium generated in fabrication processes that keep these elements isolated from one another and are free of fission product contamination.

Current studies and conceptual process and facility designs involve both reprocessing and coprocessed uranium and plutonium fuels. These material forms and compositions pose a whole new set of measurement problems, which we have only begun to address. In the international arena, the fact that fuel reprocessing and development of the breeder are proceeding now makes solving the associated measurement and accounting problems an urgent need. Some of the materials for which NDA measurement techniques should be developed, tested, and evaluated are: leached spent-fuel hulls and other "hot" scrap and waste, mixed uranium and plutonium solutions with the various levels of fission product contamination characteristic of reprocessing, "cold" mixed plutonium and uranium solutions and solids characteristic of co-converson (from nitrate to oxide), and final breeder and (perhaps plutonium recycle) fuels. Futhermore, until commercial (LWR) spent fuels are reprocessed, they must be stored at the reactor sites or at special away-from-reactor (AFR) facilities. The spent fuels should be verified and measured quantitatively, if possible, to establish their economic value and to control batch make-up for future reprocessing, as well as for safeguards.

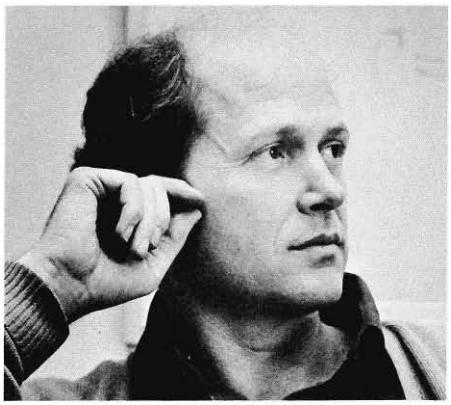
Concerns are increasing about safeguards for uranium enrichment, the front end of the fuel cycle, as advanced isotope separation processes based on centrifuges, lasers, or plasma devices are implemented. Because of their inherently large isotope separation factors, fewer separation stages are required to produce high-enriched uranium than are needed for classical gaseous diffusion separators. Hence the advanced isotope separation techniques could be more easily used for covert production of bomb materials by the facility operator. New safeguards measurement problems associated with these advanced methods must be addressed.

Process lines in future facilities will use advanced processing and control technology, including remote operation and maintenance, to minimize personnel radiation exposure, assure material containment, and handle high throughput efficiently. Almost certainly more, not less, measurement instrumentation will be needed in process lines. The instruments must function very reliably, even in such high-radiation environments as reprocessing canyons, because of the restricted access. Individual measurement stations must be designed to meet the well-ordered requirements of cost-effective integrated, automated systems of materials accounting and control. At present, LASL and the Hanford Engineering Development Laboratory are planning NDA instrumentation to be incorporated at DOE's new Fuel Materials Examination Facility at Richland in an automated mixed uranium and plutonium oxide fuel fabrication line. With the DOE/ORNL Consolidated Fuel Reprocessing Program, LASL also is working on the conceptual safeguards design for an advanced fuel-reprocessing plant.

Implementation of advanced automated materials accounting and control systems may constitute an enigma for safeguards inspectors. The question is how an inspector can independently verify that a large complex integrated system has been operated as declared. Providing system integrity with sufficient transparency for verification will undoubtedly bring changes in the design of individual instruments and their measurement controls. Inspectors will need improved portable instruments to confirm the results of the large in-plant systems.

Safeguards technology, like waste management, was once viewed by many scientists and planners as a supportive, ancillary factor in the development of nuclear power for the generation of electricity. These factors, together with reactor safety, have now become dominant in the determination of the future of nuclear energy. Thus levels of activity in future safeguards instrumentation development just described will depend to a large extent on the acceptance and use of nuclear energy, which in turn will be strongly influenced by public perception of the safety of reactors, the capabilities for safeguarding these fuels for peaceful uses, and safe management of the associated nuclear waste materials. In this chicken-and-egg cycle, we hope that the significant advances in safeguards technology, as well as in reactor safety and waste management, will be given due consideration.

THE AUTHORS



Howard O. Menlove, Group Leader of the International Safeguards Group, has worked with the Nuclear Safeguards Program at LASL for 12 years. He has been active in research and development of advanced techniques for nondestructive assay of fissionable materials. At present, his work is in inspector instrumentation development and implementation, nondestructive assay standards and calibration, spent-fuel verification techniques, training, and technology transfer. Before joining LASL, he had considerable experience in neutron and fission physics and in gamma-ray spectroscopy. After earning his Ph.D. in nuclear engineering at Stanford University, he spent a year at the Kernforschungszentrum in Karlsruhe, FRG, supported by a Fullbright Award.

Roddy B. Walton earned his bachelor of science degree in physics at Texas A&M University, and his Ph.D. in nuclear physics at the University of Wisconsin in 1957. From 1959 to 1967, he was with the General Atomic Company, where he was involved in electron linac instrumentation and experiments in neutron thermalization, neutron capture cross sections, delayed gamma rays from fission, and photonuclear reactions. In 1967, he joined LASL, and has been working in development and implementation of methods for the measurement of fissionable materials for the nuclear safeguards program. He has been instrumental in the initiation of new projects, including the Mobile Nondestructive Assay Laboratory and its operation in the field, DYMAC (dynamic materials accounting), and systems analysis. He is a Fellow of the American Nuclear Society.